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Experimental insights into the segregation of aerated bimodal particle-gas mixtures under shear

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Abstract

Gas-particle mixtures occur in numerous geophysical phenomena and industrial processes, yet describing and understanding their flow properties remains a challenge. The difficulty in modelling granular mixtures comes from having to consider the characteristics of individual particles, properties of the aerating gas, and the complex gas-particle interactions, all of which often change both temporally and spatially. Here, we present an experimental investigation into the properties of sheared monodisperse and bimodal granular mixtures at various levels of aeration. The effects of applied shear on particle segregation were analysed. At sufficiently high air velocities, monodisperse columns exhibit bubbling fluidisation, while bimodal columns segregate into a layer of coarser glass beads overlain by a layer of smaller particles. Our experimental data indicate that applied shear can suppress the formation of bubbles and promote a homogenous non-bubbling aeration in monodisperse columns, while for the bimodal columns, shear was observed to delay and suppress segregation and the column remained in a mixed state.

Keywords: fluidised bed, sheared granular flow, segregation, bubbling fluidisation

1. Introduction

Fluidised beds are particle-gas mixtures, where the solid particles are suspended within a fluid medium, enabling the granular mixture to behave like a fluid. Understanding the behaviour of fluidised beds is crucial for numerous industrial processes, where raw materials usually come in a granular form. These processes include, for example, production of pharmaceuticals [e.g., 1, 2, 3], formulation of food products [e.g., 4, 5, 6], CO₂ capture [7, 8], and utilisation of biomass alternative fuels [e.g., 9, 10, 11]. In the natural environment, enhanced understanding of particle-gas mixtures is necessary for forecasting runout distances and hazard management of geophysical flows such as snow avalanches, debris flows, and pyroclastic density currents [e.g., 12, 13, 14, 15]. Given this widespread occurrence across a range of both industrial products and natural processes, fluidised granular beds are a major area of interest within the field of granular physics [e.g., 16, 17, 18, 19, 20, 21].

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13 Experimentally, fluidised beds can be generated through the introduction of air into the base of
14 a dry granular column. When a gas flow rate is sufficiently high, the total weight of the particles
15 is balanced by the upward drag force exerted by the gas passing through the column. The internal
16 friction of the particles is reduced and the column becomes fluidised [22]. The specific value of the
17 gas velocity at which the column becomes fluidised is known as the minimum fluidisation velocity,
18 u_{mf} . At gas velocities above u_{mf} , air bubbles may form at the base of the granular column and the
19 column may exhibit bubbling fluidisation [22, 12, 23]. Previous studies show that if the granular
20 column is sheared whilst being fluidised, shear might suppress the bubble formation and promote
21 homogenous, non-bubbling fluidisation [12, 24, 23].

22
23 In many scenarios both in nature and industry, a perfectly homogenous, monodisperse particle
24 size and density distribution is unlikely. Furthermore, during the fluidisation process particle-
25 particle collisions generate fines in a process known as attrition [25, 26, 8, 27] and generate bimodal
26 grain size distributions [28, 29]. In these cases, when a granular mixture comprises particles of vary-
27 ing sizes and densities, the particles may segregate at certain gas velocities. Segregation of particles
28 is a process wherein particles of different sizes or densities separate from the mixture, leading to
29 a non-uniform distribution [17, 30, 21]. In bubbling fluidised columns, as the bubbles rise through
30 the column, they carry smaller, lighter particles to the column surface. The larger, heavier particles
31 sink down to the bottom, creating a vertical circulation of the particles in the axial direction of
32 the column [31, 32, 30, 21]. The non-uniform distribution and heterogeneity of segregated granular
33 columns significantly affects key properties such as the rheology and the flow patterns, which in
34 turn impact the performance of the column in aspects such as heat transfer and chemical reactions
35 [21]. It is therefore crucial to understand the mechanisms leading to particle segregation as well
36 as the segregation process. Additionally, across most industrial and natural applications granular
37 mixtures are usually dynamic phenomena, and parameters such as the shear rate can change both
38 spatially and temporally, influencing the particle segregation process.

39
40 The motivation for this work is to investigate how particle segregation is affected by the applied
41 shear and how particle segregation during aeration can change the flow properties of a granular
42 column. This study presents an experimental investigation into the dynamics of aerated bimodal
43 granular mixtures composed of spherical glass beads of different sizes. The granular mixtures are
44 simultaneously sheared and aerated with a known gas flow rate, and shear stress is simultaneously
45 measured. In this contribution, we present the analysis of the measured shear stress as well as
46 qualitative analysis of the bubble formation and the segregation processes.

47 2. Methodology

48 2.1. Particle and granular mixtures characterisation

49 The granular material used in the experiments consisted of Ballotini glass beads of diameter:
50 355-500 μm and 63-90 μm (supplied by Kuhmichel Abrasiv Limited). These two size fractions are
51 referred to by a nominal grain size as the catching sieve mesh size, 355 μm and 63 μm hereafter. For
52 the 355 μm particles, the mean diameter and the Sauter mean diameter are $d_{50} = 450 \mu\text{m}$ and $d_{32} =$
53 $458 \mu\text{m}$. For the 63 μm particles, the mean diameter and the Sauter mean diameter are $d_{50} = 68 \mu\text{m}$
54 and $d_{32} = 68 \mu\text{m}$. Both size fractions have a narrow particle size distribution with the ratio d_{50}/d_{32}
55 $= 0.98$ and 1.00 for 355 μm and 63 μm glass beads, respectively. The average values of sphericity
56 are 0.93 ± 0.00 and 0.84 ± 0.07 for 355 μm and 63 μm glass beads, respectively, where a sphericity,

57 ϑ , value of 1 equates to a perfect sphere. The particle density is $2491 \pm 3 \text{ kg m}^{-3}$ and $2480 \pm 2 \text{ kg}$
 58 m^{-3} for the $355 \mu\text{m}$ and the $63 \mu\text{m}$ glass beads. Detailed particle characterisation methods and raw
 59 data are provided in the supporting information. Given the particle size distribution, sphericity,
 60 and density of the $355 \mu\text{m}$ and $63 \mu\text{m}$ glass beads, both individual nominal size fractions will be
 61 considered as monodisperse, spherical, and of uniform density for further analysis. Thus, only when
 62 both are mixed they become bimodal in terms of size.

63
 64 Prior to their use, the glass beads were dried for 24 hours at 100°C in a laboratory oven to remove
 65 any moisture and then left to cool for 24 hours. The fluidisation and rheometry experiments were
 66 performed using two monodisperse and four bimodal granular mixtures with an experiment input
 67 mass m of $160.00 \pm 0.05 \text{ g}$. The monodisperse granular mixtures consisted solely of the two nominal
 68 size fractions, $355 \mu\text{m}$ and $63 \mu\text{m}$, and will be referred to as M355 and M63 hereafter. The bimodal
 69 granular mixtures were composed of the two nominal size fractions with different small-to-large mass
 70 ratios of 50:50, 40:60, 25:75, 10:90, and will be referred to as B50, B40, B25, and B10, respectively.

71 *2.2. Experimental apparatus*

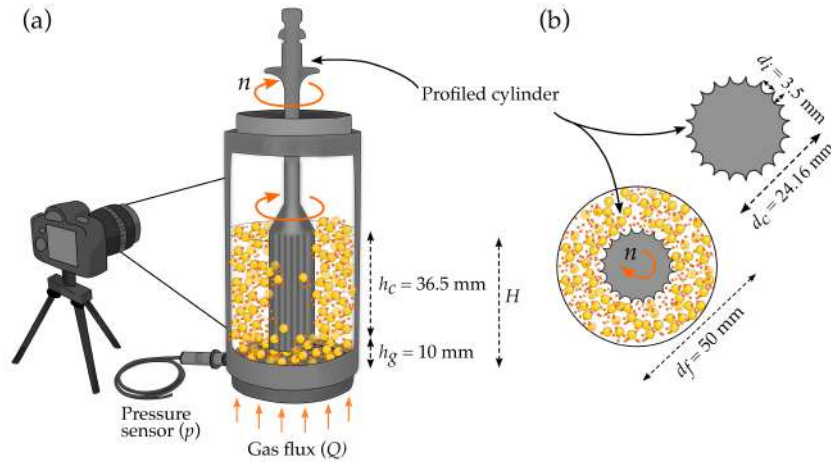


Figure 1: Diagram of the experimental apparatus modified from [23]. (a) The powder flow cell as viewed from the side; (b) The cross-section of the flow cell. Subscripts f, c, and i refer to the dimensions of the flow cell, the cylinder, and the cylinder indentations, respectively. The indentations are half-circles with a diameter of d_i . Subscript g refers to the gap set by the rheometer between the cylinder and the bottom of the flow cell.

72 The experiments were conducted using an Anton Paar MCR302 rheometer with a Powder Flow
 73 Cell attached (Figure 1). The Powder Flow Cell is a 50 mm in diameter cylindrical cell with a
 74 porous base connected to a compressed air supply to aerate the granular material. The inner cell
 75 walls are FTO-coated glass (Fluorine-doped Tin Oxide) to minimise the electrostatic effects. The
 76 compressed air is regulated by a Bürkert Mass Flow Controller operating at a range of $0\text{-}10 \text{ L min}^{-1}$
 77 for the experiments using $63 \mu\text{m}$ glass beads and all the bimodal mixtures and at a range of $0\text{-}80 \text{ L}$
 78 min^{-1} for the experiments using $355 \mu\text{m}$ particles. The pressure at the base of the flow cell is
 79 recorded by a Keller pressure sensor located at the bottom of the flow cell (with a measuring range
 80 of $800\text{-}2000 \text{ kPa}$ with a maximum resolution of 0.1 kPa).

81

82 The granular material is sheared using a profiled cylinder measuring geometry that rotates at n
 83 rotations per second. The profiled cylinder is 24.16 mm in diameter with 20 evenly spaced concave
 84 indentations, which are 3.5 mm in diameter (Figure 1). The indentations extend the entire length
 85 of the cylinder, as shown in Figure 1, preventing particle slip during rotation [33]. The profiled
 86 cylinder rotating at n rotations per second shears the granular material at a shear-rate, $\dot{\gamma}$

$$87 \quad \dot{\gamma} = CSR \times n, \quad (1)$$

88 where CSR is a Controlled shear-rate (CSR) geometry factor, typically determined by calibration
 89 with a Newtonian fluid of known viscosity [34, 35, 36]. Here, the profiled cylinder is based on ISO
 90 standard 3219 and has a CSR geometry factor of 16.392.

91 *2.3. Experimental procedure and data collection*

92 Bimodal granular mixtures were studied through shear stress measurements of a granular col-
 93 umn simultaneously sheared at a constant shear-rate, $\dot{\gamma}$ and aerated with a constant air velocity,
 94 u . The experiments were performed for all the bimodal mixtures (B50, B40, B25, and B10) as well
 95 as the monodisperse mixtures (M355 and M63) for reference.

96 The shear stress, τ of the granular mixtures was measured at a range of gas velocities, u . To
 97 maintain a common reference point between the different columns and the measurements collected
 98 at different u , we use the apparent minimum fluidisation velocity, u_{amf} , which was determined by
 99 performing a series of pressure drop experiments. The procedure for the pressure drop experiments
 100 was as follows. The granular material was loaded into the Powder Flow Cell and mixed thoroughly
 101 with a spatula by hand to ensure a uniform particle size distribution throughout the measurement
 102 apparatus. The compressed air supply was then connected and fed into the base of the cell to aerate
 103 the glass beads. The glass beads were aerated with an increasing air flow rate, Q , from 0 L min^{-1}
 104 to Q_{max} , while the rheometer and the associated software collected the pressure data, p . The value
 105 of Q_{max} was 30, 1.5, 1.7, 1.0, 4.0, 8.0 L min^{-1} for the M355, M63, B50, B40, B25 and B10 mixture,
 106 respectively. The flow rate values were converted to air velocity using the cross-sectional area of the
 107 flow cell. The same experiments were performed with an empty Powder Flow Cell to obtain a refer-
 108 ence value of the pressure, p_{air} , for each Q_{max} , i.e., for each granular mixture. The pressure drop
 109 across the granular column was then calculated as $\Delta p = p - p_{air}$. The apparent minimum fluidi-
 110 sation velocity, u_{amf} was determined from visual inspections of the pressure drop profiles produced.
 111

112 The experimental procedure of the stress measurements was as follows. The granular material
 113 was loaded into the Powder Flow Cell and mixed thoroughly with a spatula by hand, again to
 114 ensure a uniform particle size distribution throughout the measurement apparatus. Although the
 115 glass beads are nearly perfectly spherical, the granular mixture was pre-sheared at $n = 1 \text{ rps}$ for
 116 pre-shear time $t_{ps} = 180 \text{ s}$ prior each measurement. This ensured that the particles have established
 117 the equilibrium orientation distributions and minimised transient oscillations in the apparent shear
 118 stress [37]. We refer the reader to Figure S3 in the supporting information which presents the stress
 119 measurements for a range of t_{ps} .
 120

121 At time $t = 0 \text{ s}$, the compressed air supply was turned on to aerate the glass beads with a con-
 122 stant flow rate of $u = 0 u_{amf}$, $0.5 u_{amf}$, $1.0 u_{amf}$, $1.5 u_{amf}$ and $2.0 u_{amf}$. The granular column was
 123 also simultaneously sheared by the profiled cylinder rotating at a constant rate of n rotations per
 124 second, $n = 0.5, 1, 3, 5, 7, 9, 13 \text{ rps}$, which corresponds to $\dot{\gamma} = 8, 16, 49, 82, 115, 148, 213 \text{ s}^{-1}$. The
 125

126 rheometer and the associated software collected the dataset, which included the shear stress, τ and
 127 the time step between the data points, dt . One hundred data points were collected, corresponding
 128 to a specific shear stress, and each individual data point was captured when the shear stress reached
 129 a stable value.

130
 131 Alongside data recorded by the rheometer, additional data were also collected using an Olympus
 132 OM-D E-M10 Mark IV camera. The experiments were recorded at 2160×3840 pixels resolution at
 133 30 frames per second with a 14-42 mm lens. The camera was located at the side of the rheometer
 134 and focused on the transparent flow cell window to capture the granular column. The videos were
 135 inspected visually to observe the aeration and segregation processes. Individual video frames were
 136 processed in MATLAB[®] to manually extract the height of the granular column, H . Having these
 137 data, the particle volume fraction, ϕ was then calculated from

$$138 \quad \phi = \frac{(r_1 \times m)/\rho_1 + (r_2 \times m)/\rho_2}{A_f h_g + A_p(H - h_g)}, \quad (2)$$

139 where m is the mass of the granular mixture, ρ_1 , ρ_2 , r_1 and r_2 are the density and the percent-
 140 age ratio of the $355 \mu\text{m}$ and $63 \mu\text{m}$ glass beads, respectively, A_f is the area of the base of the
 141 flow cell, A_p is the area of the ring around the profiled cylinder occupied by the particles and h_g
 142 is the gap set by the rheometer between the profiled cylinder and the base of the flow cell (Figure 1).
 143

144 Individual video frames of the experiments where segregation was observed were also processed
 145 in MATLAB[®] to extract the temporal evolution of the thickness of the layer of $63 \mu\text{m}$ particles, h_f .
 146 These data were then analysed to determine the timescale of segregation, t_{seg} , as the time where
 147 the $63 \mu\text{m}$ layer reached its maximum thickness.

148 3. Results and discussion

149 3.1. Minimum fluidisation velocity

150 Figure 2 shows the pressure drop, Δp , across the granular column as a function of air velocity,
 151 u , for all the studied granular mixtures. For all the granular columns, Δp increases linearly with
 152 increasing u until it reaches a maximum value. For the monodisperse columns, after Δp reaches the
 153 maximum value, it plateaus. This occurs at the point when the weight of the column is balanced
 154 by the drag imposed by the upward gas flux and the column is fluidised. However, for the bimodal
 155 columns the behaviour is different, after Δp reaches the maximum value, it rapidly decreases and
 156 reaches a somewhat steady value. From the visual inspection of videos of the experiments, we
 157 suggest that Δp begins to decrease as the granular columns begin to segregate into separate $355 \mu\text{m}$
 158 and $63 \mu\text{m}$ layers. The somewhat constant and steady value of Δp corresponds to the time, and
 159 hence the air velocity, when the column has reached a stable segregated state. For consistency, we
 160 determine an apparent minimum fluidisation flow rate as the value of u at which Δp reaches the
 161 maximum value for all the mixtures. The apparent minimum fluidisation velocity was thus deter-
 162 mined to be $u_{amf} = 0.168, 0.006, 0.004, 0.005, 0.025, 0.036 \text{ m s}^{-1}$ for the granular mixtures M355,
 163 M63, B50, B40, B25 and B10, respectively. The experimentally determined apparent minimum
 164 fluidisation velocity will be referred to as u_{amf} , while the theoretical value and the concept of the
 165 minimum fluidisation velocity will be referred to as u_{mf} hereafter.
 166

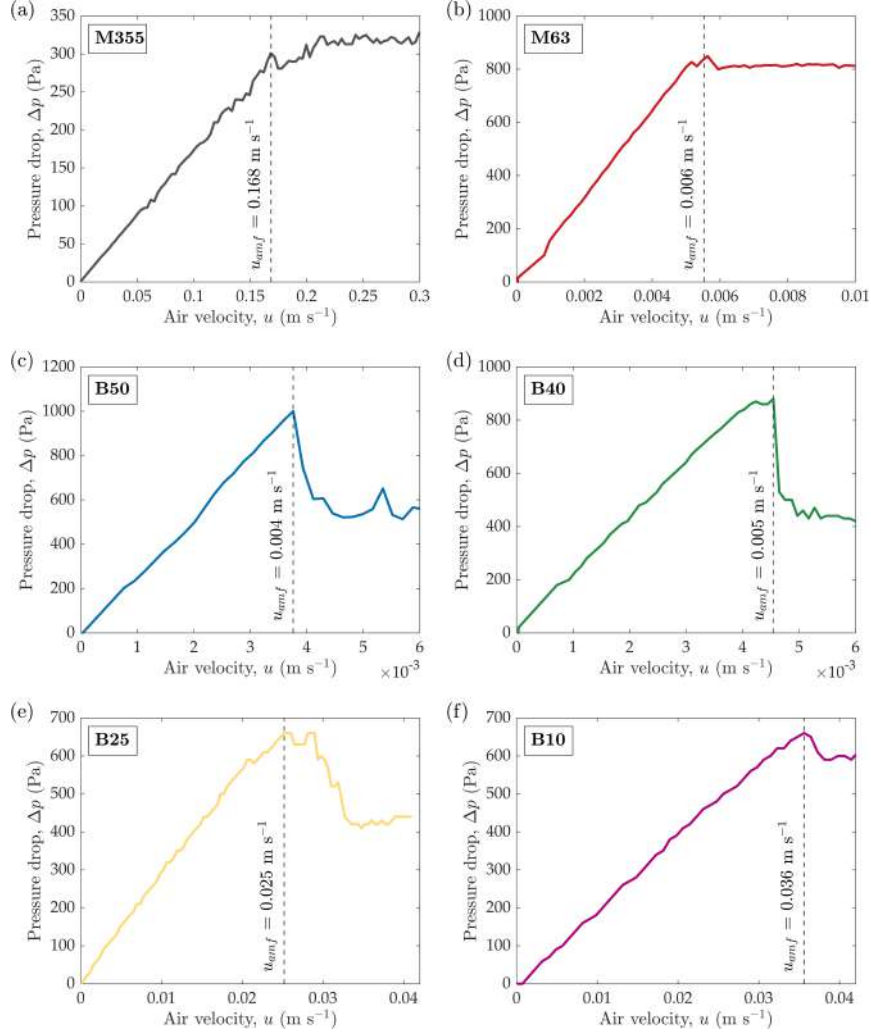


Figure 2: Pressure drop across the granular column as a function of the air flow rate under for the two monodisperse granular mixtures, (a) M355, (b) M63, and the four bimodal granular mixtures, (c) B50, (d) B40, (e) B25, (f) B10. The dashed lines represent apparent minimum fluidisation velocity, u_{amf} .

167 Although our method of determining u_{amf} was consistent for all the mixtures, it became ap-
 168 parent that there are significant challenges in determining the minimum fluidisation velocity of
 169 bimodal granular mixtures due to the large and small particles being fluidised at different velocities
 170 and hence the segregation of the granular column into separate 355 μm and 63 μm layers. Although
 171 the density ratio of the two nominal particle fractions is $\rho_1/\rho_2 \sim 1$, the size ratio is $d_1/d_2 \sim 7$,
 172 where the subscripts 1 and 2 correspond to the large and small particles, respectively. Consequently,
 173 since u_{mf} strongly depends on the particle size, the 355 μm glass beads require 30 times greater air
 174 velocities to become fluidised than the 63 μm particles, $u_{mf1}/u_{mf2} \sim 30$.

175

176 We may estimate that the value of the minimum fluidisation velocity of a bimodal mixture
 177 comprising 355 μm and 63 μm glass beads should fall between the value of minimum fluidisation
 178 velocity for 63 μm and 355 μm particles, i.e., $0.006 \text{ m s}^{-1} = u_{mf2} < u_{mf} < u_{mf1} = 0.168 \text{ m s}^{-1}$
 179 [38, 21]. However, the experimentally determined values of u_{amf} for B50 and B40 are lower than
 180 u_{mf} for 63 μm , suggesting that the segregation of particles have significantly affected the pressure
 181 drop experiments and, in general, introduces uncertainties when determining u_{mf} for bimodal mix-
 182 tures. As soon as the column is subjected to an air velocity greater than the minimum fluidisation
 183 velocity, the 63 μm particles move in-between the 355 μm particles and migrate towards the top of
 184 the column. It is therefore difficult to experimentally determine the exact value of u_{mf} for bimodal
 185 mixtures comprising particles of similar densities but with large size ratio.

186
 187 The pressure drop method, as used here, was applied to experimentally determine the minimum
 188 fluidisation velocity of bimodal mixtures by several previous studies [e.g., 39, 40, 41, 38, 42, 20].
 189 These data have then been used to propose semi-empirical relationships between u_{mf} and the
 190 parameters of the bimodal mixtures. For example, [20] modified the relationship by Cheung et al.
 191 [40] and put forward the equation

$$192 \quad u_F = u_2 \left(\frac{u_1}{u_2} \right)^{v^\kappa}, \quad (3)$$

193 with

$$194 \quad v = r_1 \frac{\bar{\rho}}{\rho_1}, \quad (4)$$

$$195 \quad \kappa = 1.26 \left(\frac{d_1}{d_2} \right)^{0.53}, \quad (5)$$

196 and the average particle density defined as

$$197 \quad \frac{1}{\bar{\rho}} = \frac{r_2}{\rho_2} + \frac{r_1}{\rho_1}, \quad (6)$$

198 where the subscripts 1 and 2 correspond to the large and small particles, respectively. However,
 199 several authors have argued that a bimodal mixture cannot be defined with one specific value of
 200 minimum fluidisation velocity because the fluidisation takes place along an extended velocity inter-
 201 val with boundaries defined as the *initial*, u_{imf} and the *final fluidisation velocity* of the mixture,
 202 u_{fmf} [43, 44, 45, 21]. This theory suggests that if $u > u_{imf}$, the granular column segregates into
 203 three layers: a bottom mixed region, a layer of large particles, and a bubbling layer of small particles
 204 located at the top of the column. As the air velocity increases to $u > u_{fmf}$, the bubbles travel
 205 vertically throughout the whole column and the column becomes mixed again. Further investiga-
 206 tion into these theories is beyond the scope of this study. Here, we use the apparent minimum
 207 fluidisation velocity simply as a reference point to relate the experiments performed using different
 208 mixtures and at different air velocities.

209
 210 Alongside the u/u_{amf} ratio, each value of u corresponds to a different particle volume fraction,
 211 ϕ , which can be calculated from the image analysis of our recorded videos. This relationship is
 212 shown in Figure 3. In our experiments, for all the granular mixtures, as u increases ϕ becomes
 213 lower or equal to the initial particle volume fraction, i.e., the particle volume fraction, ϕ_0 for u

214 $= 0 \text{ m s}^{-1}$. Therefore, we are confident that, in our experiments, all the granular mixtures are
 215 in different states of aeration for different u/u_{amf} . These results also highlight that a particle
 216 volume fraction, in particular, in relation to the initial volume fraction, should be considered when
 217 describing different fluidisation and aeration regimes. We will refer to the monodisperse columns
 218 as fluidised and to bimodal columns as aerated hereafter.

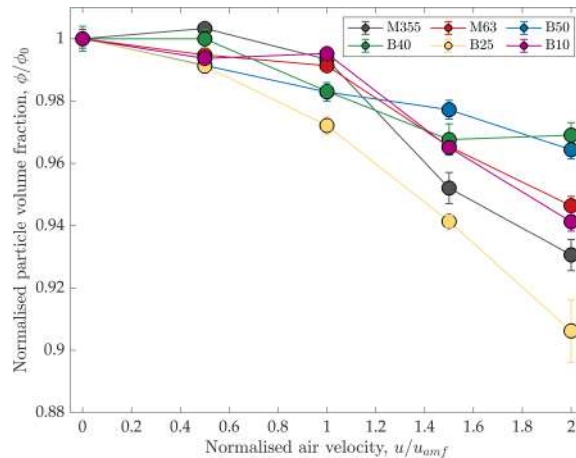


Figure 3: Normalised particle volume fraction, ϕ/ϕ_0 , as a function of normalised air velocity, u/u_{amf} . The different coloured data represent different granular mixtures. The lines are not model curves and are simply used to guide the eye.

219 3.2. Experimental observations

220 When the air flow is introduced into the base of a granular column, the column starts to ex-
 221 pand vertically and increases in height, which consequently reduces the particle volume fraction, ϕ .
 222 Column expansion is observed for all the granular columns, all the shear-rates, $\dot{\gamma}$, and all the flow
 223 rates $u > 0 \text{ m s}^{-1}$, however it is most pronounced for $u \geq u_{amf}$. The monodisperse columns are
 224 homogeneously fluidised at $u = u_{mf}$ and experience bubbling fluidisation at $u > u_{mf}$. In bubbling
 225 fluidisation, air bubbles form at the base of the columns, move vertically upwards and most erupt,
 226 ejecting particles from the column surface. The fluidisation processes of sheared granular columns
 227 comprising monodisperse particles have been described in detail in a previous study [23]. Here, we
 228 focus on the aeration and segregation behaviour of the bimodal granular mixtures.

229 Figure 4 presents individual image frames of the experiments for the four bimodal granular
 230 columns, B50, B40, B25, and B10. While the specific images were chosen such that they present
 231 the most prominent processes and features, the observations are true for all the experiments per-
 232 formed using bimodal mixtures. When the bimodal columns are aerated with air velocities $u >$
 233 0 m s^{-1} , the $63 \mu\text{m}$ glass beads (i.e., the finer particles) begin to move towards the top of the col-
 234 umn. After a given time, the bimodal column may reach a fully segregated stable state with a
 235 coarse layer of $355 \mu\text{m}$ glass beads overlain by a thinner layer of $63 \mu\text{m}$ particles (Figures 4d and
 236 4f). The upper layer of $63 \mu\text{m}$ particles often experiences bubbling. The $355 \mu\text{m}$ glass beads require
 237 greater air velocities to be fluidised and were only observed to experience bubbling when in the
 238 mixed state with the $63 \mu\text{m}$ particles. If the energy provided by the air velocity is not sufficient or if
 239

240 it is balanced by other forces, the granular column remains in a mixed state (Figures 4a-b and 4g-h).
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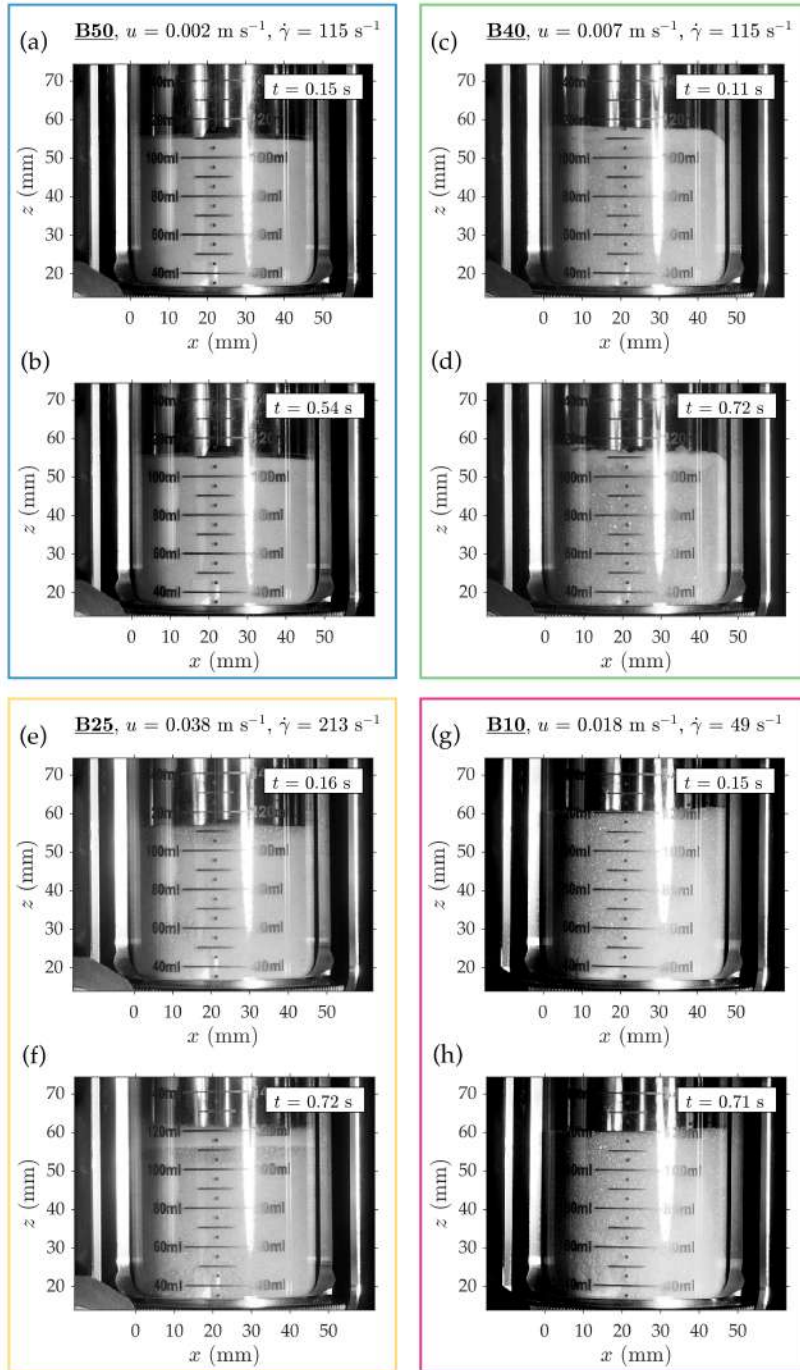


Figure 4: Individual image frames from videos of the experiments for the four bimodal columns, (a)-(b) B50, (c)-(d) B40, (e)-(f) B25, and (g)-(h) B10. The air velocity, u , and the shear-rate, $\dot{\gamma}$, is written above each experiment. The time step of each image frame is written in the top right-hand corner of each image, where $t = 0$ s is the start of the experiment. The origin of the coordinate system (x, z) is located at the bottom left corner of the flow cell.

242 A small number of the experiments exhibited processes such as uneven particle segregation with
 243 a preference for one side of the column and electrostatic interactions between the particles and the
 244 rheometer, causing the glass beads to stick to the flow cell and the profiled cylinder. It is possible
 245 that some of these processes were a result of heterogenous starting conditions, for example, the
 246 initial granular column was not mixed thoroughly before the start of the experiment and the par-
 247 ticles were not evenly distributed. However, it is also possible that these processes are equipment-
 248 and experiment-specific. These experiments constitute approximately 10% of all the experiments
 249 performed using bimodal mixtures and it was not possible to process those videos to extract reliable
 250 quantitative data.

252 3.3. The effects of shear on column segregation

253 We illustrate the effects of shear on the segregation of the bimodal granular columns through
 254 image analysis of the captured videography. Figure 5 shows individual frames of the B25 granular
 255 column aerated at $u/u_{amf} = 1$ and sheared at $\dot{\gamma} = 16 \text{ s}^{-1}$. Similarly, to the monodisperse granular
 256 columns, as the air velocity increases above the minimum fluidisation velocity, bubbles start to form
 257 at the base of the bimodal column and they travel vertically upwards to the column surface (Figure
 258 5a-b). The bubbles are of different sizes and they are not necessarily spherical, as shown in Figure
 259 5b. As the bubbles rise through the column, they carry a wake of smaller particles to the top of
 260 the column. This process can be seen in Figure 5c-d, where the green arrow indicates a bubble that
 261 travels from approximately $z = 50 \text{ mm}$ to the column surface. At the surface (in Figure 5d), the
 262 breaching bubble is surrounded by a boundary composed of smaller particles. While the smaller
 263 particles ascend with the bubbles, the larger, heavier particles sink to the bottom of the column.
 264 This results in an overall circulation of particles in the axial direction of the column. The bimodal
 265 column thus begins to segregate into a layer of larger and smaller particles, a layer of $355 \mu\text{m}$ glass
 266 beads and a layer of $63 \mu\text{m}$ glass beads in our study. Given that the $63 \mu\text{m}$ glass beads are nearly
 267 seven times smaller than the $355 \mu\text{m}$ glass beads and become fluidised at much smaller air velocities,
 268 the $63 \mu\text{m}$ glass beads may also be travelling vertically upwards in the spaces between the larger
 269 particles, contributing to the segregation process. While still partly mixed, the granular column
 270 in Figure 5e shows a greater proportion of small particles in its upper layers. The bubbles are
 271 still present, indicating that the particle circulation, and hence the column segregation, is still in
 272 progress. Figure 5e shows a segregated granular column with a layer of $355 \mu\text{m}$ glass beads overlain
 273 by a layer of $63 \mu\text{m}$ glass beads. While the coarser layer is likely to still contain some proportion of
 274 small particles, the upper layer is composed of the small particles only.

275
 276 The series of images in Figure 5 shows a granular column subjected to the normalised air ve-
 277 locity, $u/u_{amf} = 1$. As previous research suggests, it is likely that at greater air velocities both the
 278 smaller and the larger particles would fluidise and the column would remix again [43, 44, 45, 21].
 279 In our study, some of the experiments conducted at greater air velocities exhibited electrostatic in-
 280 teractions between the particles and the rheometer cell walls. The glass beads stuck to the flow cell
 281 and the profiled cylinder, obstructing the view of the rest of the column. It is therefore likely that
 282 greater air velocities were sufficient to fluidise and remix both the smaller and the larger particles,
 283 but visual confirmation was obstructed.

284
 285 Previous research shows that the onset of bubbling in monodisperse columns can be suppressed
 286 by applying shear to the granular column [12, 24, 23]. If the power input from shear is greater

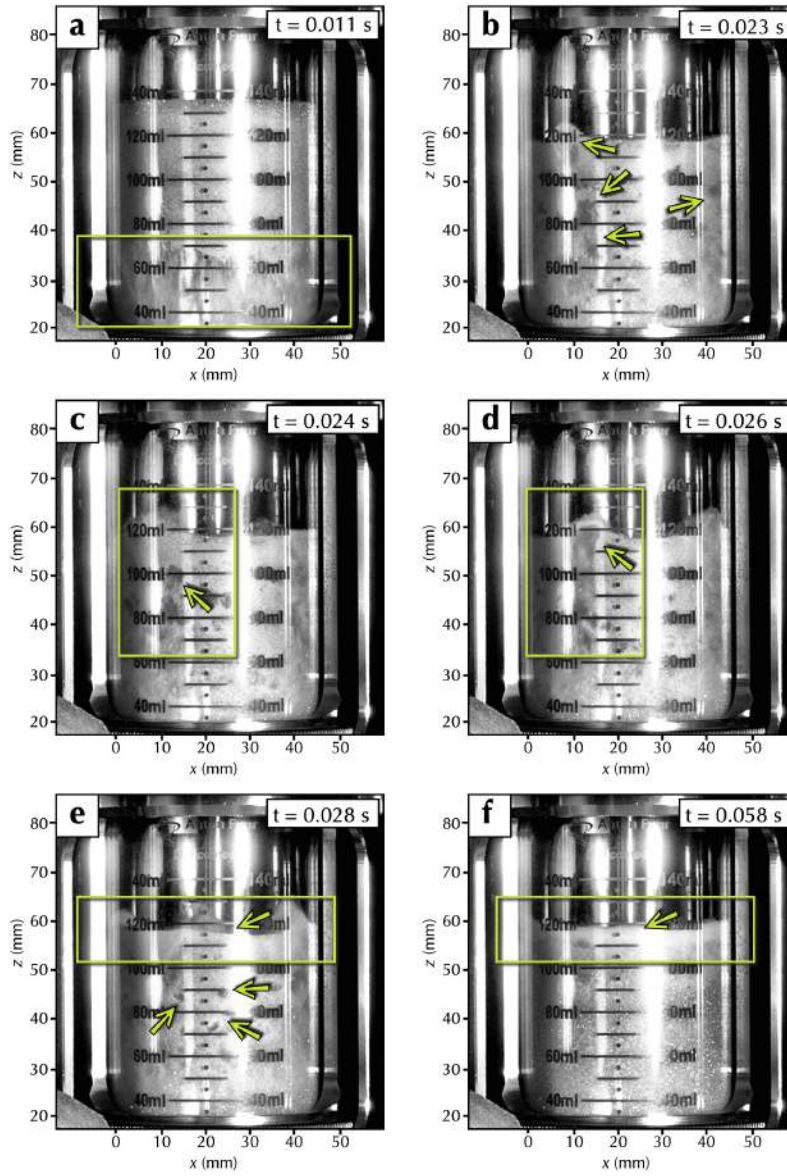


Figure 5: Individual image frames showing how the B25 granular column evolves in time (a)-(f) $t = 0.011$ s to 0.058 s whilst aerated at normalised air velocity, $u/u_{amf} = 1$ and sheared at $\dot{\gamma} = 16 \text{ s}^{-1}$. The origin of the coordinate system (x, z) is located at the bottom left corner of the flow cell. The green arrows and boxes point to air bubbles and other features discussed in the main text.

287 than the power provided by the air flow, any bubbles that may form at the base of the column are
 288 suppressed and the column remains homogeneously (non-bubbling) fluidised. This indicates that
 289 there are two regimes: a fluidisation-dominated regime, where the fluidisation power is greater than

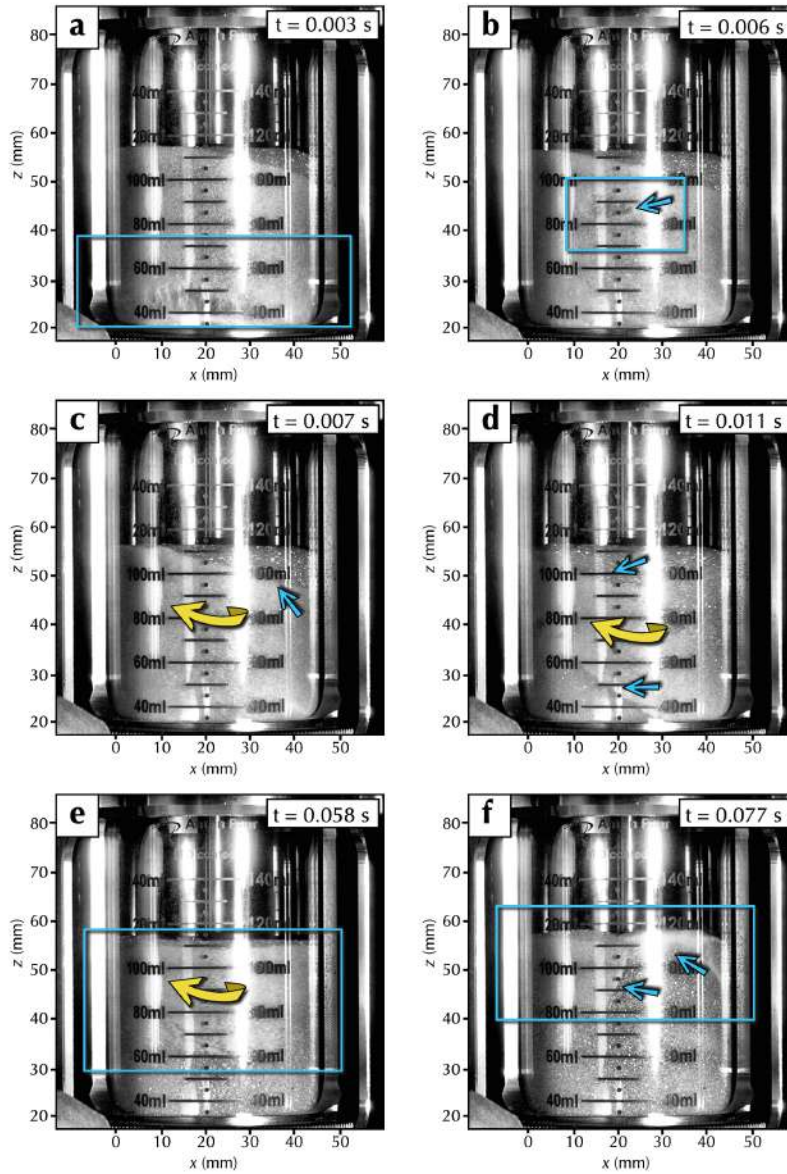


Figure 6: Individual image frames showing how the B25 granular column evolves in time (a)-(f) $t = 0.011$ s to 0.058 s whilst aerated at normalised air velocity, $u/u_{amf} = 1$ and sheared at $\dot{\gamma} = 213 \text{ s}^{-1}$. The origin of the coordinate system (x, z) is located at the bottom left corner of the flow cell. The yellow arrows indicate the movement of the particles and the air bubbles, while the blue arrows point to features discussed in the main text.

290 the shear, and a shear-dominated regime, where the shear-rate power is greater than the fluidisation
 291 input [46]. The power balance between fluidisation (aeration) and shear-rate becomes crucial in
 292 bimodal granular columns because it can indicate if a mixture will segregate at a given value of

293 shear-rate and air velocity.

294

295 When the air is introduced into the base of the granular column, the fluidisation power, P_D ,
296 results in an increase in the velocity of the particles. Because the column is dense (with an average
297 particle volume fraction of $\phi \sim 0.67$), an increased particle velocity leads to increased particle-
298 particle and particle-wall collisions, which in turn can lead to a decrease in the particle velocity.
299 Similarly, the applied shear power, $\tau\dot{\gamma}$, can affect the velocity of the particles. The average velocity
300 fluctuations of particles in a fluidised granular column are often quantified using the granular
301 temperature, T [47, 48]. The power balance between fluidisation and shear can then be expressed
302 as

303

$$\rho P_D + \tau\dot{\gamma} = \rho\Gamma\omega_c(T)T, \quad (7)$$

304 where ρ is the average bulk density of the granular column, Γ is the dimensionless dissipation
305 rate related to the coefficient of restitution and ω_c is the frequency of collisions [49, 46, 50]. The
306 fluidisation power is usually calculated by considering the granular temperature and the collision
307 frequency of the granular system [46], both of which depend on particle size and particle size dis-
308 tribution, i.e., degree of polydispersity [51, 52]. While the segregation in granular mixtures with a
309 smaller particle size ratio than investigated here might be suppressible under very different shear
310 rates, we suggest that the nondimensional quantities governing the power balance might be similar.
311 Therefore, although our experimental dataset does not provide sufficient information for detailed
312 analysis of the granular temperature, we suggest that further investigation into nondimensional
313 quantities governing fluidisation and shear power might be key to predicting the transition point
314 between mixed and segregated states. The power balance will now be discussed using examples
315 from our experiments.

316

317 Figure 6 shows individual frames of the same granular column, B25, aerated with the same
318 normalised air velocity, $u/u_{amf} = 1$, however this column now is sheared at $\dot{\gamma} = 213\text{ s}^{-1}$. Initially,
319 we observe similar features as in Figure 5 for $\dot{\gamma} = 16\text{ s}^{-1}$. As the air velocity increases, bubbles
320 start to form at the base of the column and they rise through the column to its surface. However,
321 it quickly becomes apparent that the energy provided by the upward air velocity is competing
322 with the energy input from shear. The bubbles try to carry the smaller particles to the top of
323 the column, whilst the whole column is being sheared and rotates in the clock-wise direction, as
324 indicated by the yellow arrows in Figure 6c-d. It appears that the vertical component of the ve-
325 locity of the smaller particles increases with the rising bubbles while the radial component of the
326 velocity depends on the applied shear. Consequently, the small particles are moving in a spiral-
327 like pattern, as indicated by the blue arrows pointing to the edges of a wide strip of smaller particles.

328

329 Although Figure 5f and Figure 6e show the granular column at the same time, $t = 0.058\text{ s}$, there
330 are clear differences between the two images. The granular column in Figure 5f shows a layer of
331 larger particles and a layer of smaller particles. On the other hand, the column in Figure 6e is still
332 rotating and in the process of segregation. More than half of this column still contains a significant
333 proportion of small particles mixed with the large particles. A somewhat segregated column is
334 shown in Figure 6f, where we observe a layer of large particles at the bottom and a partially mixed
335 upper part, consisting of a mixed sublayer and a thin sublayer of small particles at the column
336 surface.

337

338 Interestingly, the mixed sublayer is not uniformly mixed in the radial direction. Previous re-
 339 search shows that, particles adjacent to the profiled cylinder have a rotational velocity equal to
 340 the rotational velocity of the profiled cylinder [53]. The velocity of the particles decays in the
 341 radial plane towards the sides of the flow cell and the particles adjacent to the sides of the flow cell
 342 are nearly stationary. This leads to a heterogeneous particle volume fraction in the radial plane
 343 [53, 54, 55, 56, 23]. In bimodal granular mixtures, this may result in heterogeneities in the mix-
 344 ing/segregation in the radial plane. It may be easier for small particles to travel vertically upwards
 345 to the column surface in the slightly less-dense area around the profiled cylinder than for particles
 346 in the more-dense area close to the sides of the flow cell. This may be a reason for the observed
 347 rotating strip of small particles adjacent to the sides of the cell in Figure 6c-d. If the flow cell was
 348 wider, the applied shear might not be sufficient to cause the column rotation and might result in
 349 a (thick) stationary layer of particles adjacent to the sides of the cell. Conversely, particles in the
 350 area around the profiled cylinder have greater rotational velocities, which, in theory, should be able
 351 to balance the air velocities and remain in the mixed state. It is, however, important to note that
 352 all the observations are based on 2D images of 3D processes.

353
 354 Nevertheless, this analysis highlights the complexity of processes governing particle segregation
 355 and illustrates that when a fluidised (aerated) granular column is subjected to shear, shear affects the
 356 internal dynamics. Analogously to previous research conducted on monodisperse granular mixtures
 357 [12, 24, 23], our experiments show that the onset of bubbling can be suppressed by applying shear to
 358 the granular column. Sheared monodisperse columns exhibit non-bubbling fluidisation over a wider
 359 range of air velocities. In this study, shear delays bubble formation in bimodal granular columns,
 360 effectively resulting in mixed-state columns over a wider range of air velocities. In general, if the
 361 power input from shear is greater than the fluidisation (aeration) power, bubble formation and
 362 hence the segregation is suppressed, and the column remains in a mixed or a partially mixed state.

363 3.4. Shear stress measurements: mixed and segregated states

364 To further investigate the effects of shear on bimodal granular columns, we analyse the shear
 365 stress data collected by the rheometer. Figure 7 presents measurements of the shear stress, τ as
 366 a function of time, t of monodisperse granular mixtures M355 and M63 for a range of shear and
 367 air flow rates. As expected, apart from very minor oscillations, the shear stress is constant in time
 368 for a constant shear-rate, for both M355 and M63 mixtures for all the studied shear-rates and air
 369 velocities. For both mixtures, the shear stress is lower when the granular columns are fluidised with
 370 greater air velocities, i.e., for lower particle volume fractions, which means that it is easier for the
 371 aerated ($u > 0 \text{ m s}^{-1}$) granular mixtures to start flowing than for a dry, dense ($u = 0 \text{ m s}^{-1}$) gran-
 372 ular material. In accordance with the present results, previous studies have demonstrated that the
 373 apparent viscosity of fluidised granular mixtures decreases with the greater air velocities, enhanc-
 374 ing the mobility of the mixture [e.g., 57, 58, 59, 60]. Similarly, since granular mixtures M355 have
 375 greater particle volume fraction than M63 mixtures, the shear stress is greater for M355 columns
 376 than for M63 when fluidised with the same normalised air velocity, u/u_{amf} and sheared at the
 377 same shear-rate, $\dot{\gamma}$. These result suggests that granular columns comprising $63 \mu\text{m}$ glass beads are
 378 more mobile than columns composed of $355 \mu\text{m}$ particles and is consistent with previous research
 379 which found that the apparent viscosity of fluidised monodisperse granular mixture increases with
 380 higher particle volume fraction [e.g., 58, 61]. Additionally, our results show that the shear stress of
 381 both mixtures depends on the applied shear-rate (compare Figure panels 7a with 7d), indicating

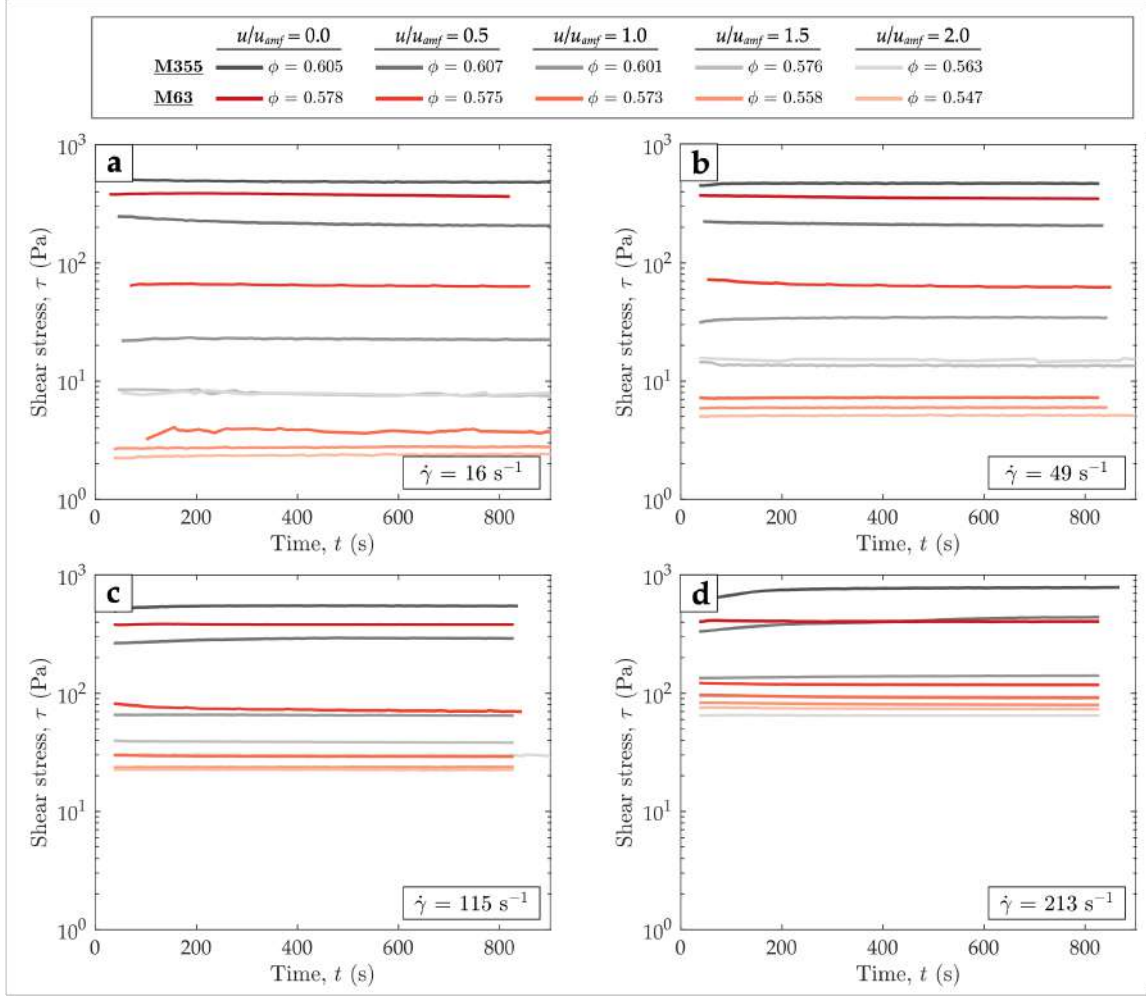


Figure 7: Shear stress, τ , of the monodispersed granular mixtures, M355 and M63, as a function of time, t , fluidised with a constant air velocity, u , and sheared at a constant rate, (a) $\dot{\gamma} = 16 \text{ s}^{-1}$, (b) $\dot{\gamma} = 49 \text{ s}^{-1}$, (c) $\dot{\gamma} = 115 \text{ s}^{-1}$, and (d) $\dot{\gamma} = 213 \text{ s}^{-1}$. The particle volume fraction, ϕ , of the granular mixtures fluidised at various air velocities is provided in the legend at the top of the figure. The y -axis, shear stress, τ , limits are the same on all panels to support cross-comparison.

382 non-Newtonian rheological behaviour [57, 58, 36, 33].

383

384 The measurements of τ , of the bimodal granular mixtures B50, B40, B25, and B10 for a range
 385 of air flow and shear-rates are shown in Figure 8. We distinguish three types of experiments: (i)
 386 experiments where no segregation was visually observed and the granular columns remained in a
 387 mixed state for the duration of the experiment (solid lines in Figure 8), (ii) experiments where clear
 388 segregation was observed and the column segregated into a layer of 355 μm and 63 μm glass beads
 389 (dashed lines in Figure 8), and (iii) experiments which exhibit partial, uneven, partial segregation

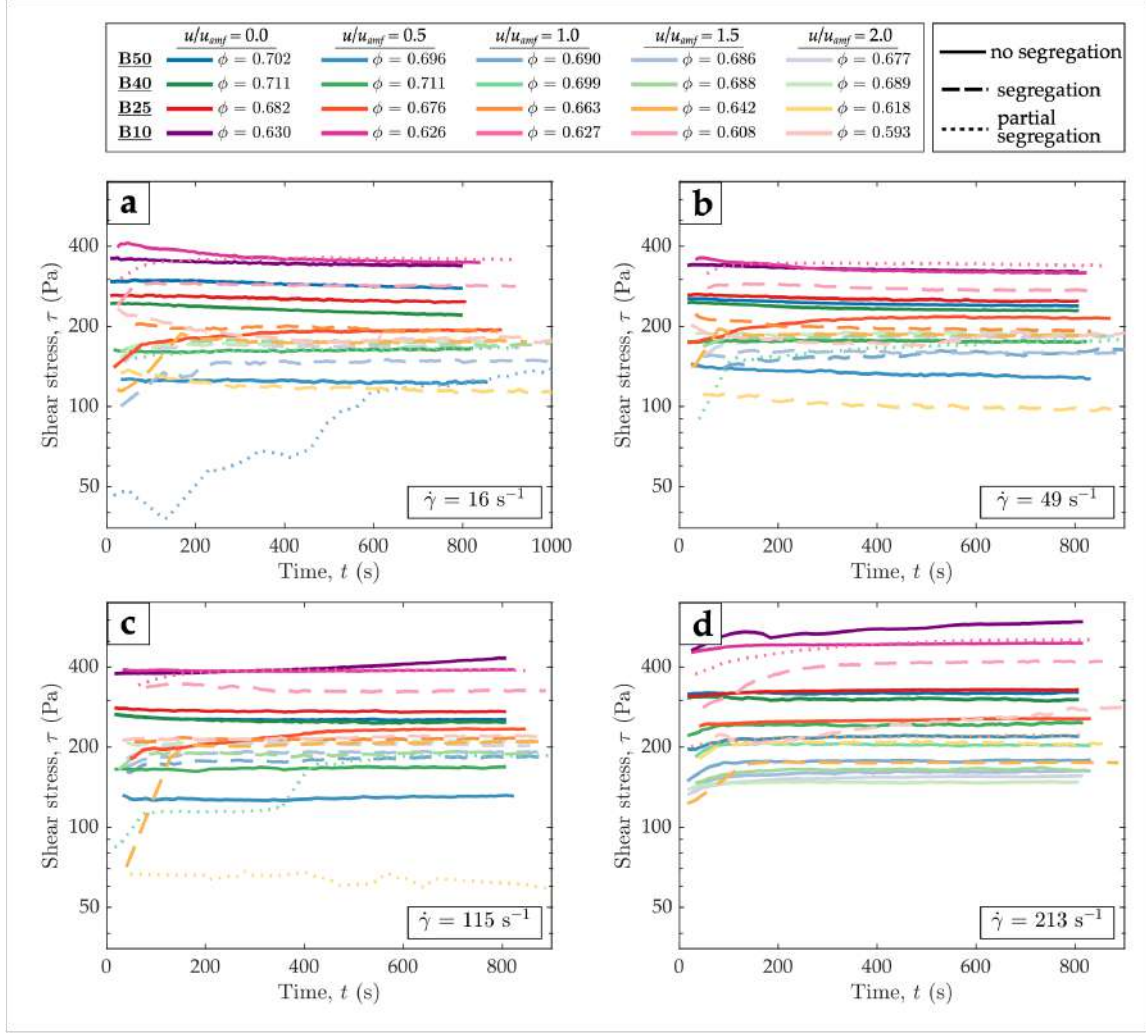


Figure 8: Shear stress, τ , of the bimodal granular mixtures, B50, B40, B25, and B10, as a function of time, t , fluidised with a constant air velocity, u , and sheared at a constant rate, (a) $\dot{\gamma} = 16 \text{ s}^{-1}$, (b) $\dot{\gamma} = 49 \text{ s}^{-1}$, (c) $\dot{\gamma} = 115 \text{ s}^{-1}$, and (d) $\dot{\gamma} = 213 \text{ s}^{-1}$. The solid lines represent experiments where no segregation was observed, the dashed lines represent experiments where segregation was clearly observed, and the dotted lines represent experiments which were discarded in further analysis. The particle volume fraction, ϕ , of the granular mixtures fluidised at various air velocities is provided in the legend at the top of the figure. The y -axis, shear stress, τ , limits are the same on all panels to support cross-comparison.

390 or other unusual features (dotted lines in Figure 8).

391

392 Regardless of the shear-rate applied, all the bimodal mixtures remain in the mixed state for
 393 the air velocities lower than the apparent minimum fluidisation velocity, $u < u_{amf}$. Conversely, in
 394 our experiments, whether the segregated state is observed for $u \geq u_{amf}$ depends on the applied
 395 shear-rate. The segregation process also appears to be controlled by the particle size distribution

396 (i.e., the degree of bimodality) because not all the bimodal mixtures achieve a segregated state for
397 the same shear-rate and the same normalised air velocity.

398
399 Similarly to the monodisperse columns, for all the shear-rates, the shear stress of the majority
400 of the bimodal mixtures in the mixed state is relatively constant in time. However, we observe an
401 initial increase in shear stress for some of the mixtures, e.g., B25 mixture aerated at $u = 0.5u_{amf}$
402 and sheared with $\dot{\gamma} = 16$ and 49 s^{-1} . Although all the mixtures were pre-sheared before the mea-
403 surements, the initially lower shear stress values could be attributed to the particles rearranging
404 themselves into a stable configuration state for the given shear-rate and flow rate. The timescale of
405 this rearrangement is most likely slightly greater than the time required for initial data acquisition.

406
407 While shear stress of the bimodal mixtures in the segregated state also increases until it reaches
408 a plateau (dashed lines in Figure 8), the timescales are longer, and these data correspond to dif-
409 ferent processes. The analysis of the data collected by the rheometer alongside the videos of the
410 experiments indicates that here, the shear stress values reflect the evolving configuration of the
411 granular mixture, where the particles are rearranging themselves to form a stable segregated state
412 with $63\text{ }\mu\text{m}$ glass beads on top of $355\text{ }\mu\text{m}$ particles.

413
414 Figure 9a presents an example dataset extracted from individual video frames, specifically, the
415 thickness of the $63\text{ }\mu\text{m}$ layer, h_f , as a function of time, t are reported. Although there are differences
416 in the values of h_f and t between the experiments, this figure and the data pattern are represen-
417 tative of all the bimodal mixtures that segregated into $355\text{ }\mu\text{m}$ and $63\text{ }\mu\text{m}$ layer. As the granular
418 column is subjected to a sufficiently high air flow h_f increases until it reaches a relatively stable
419 maximum value at time $t = t_{seg}$. The upper $63\text{ }\mu\text{m}$ layer often exhibits bubbling, which results in
420 fluctuations of h_f about the plateau. The values of t_{seg} for all the bimodal granular columns for
421 a range of air velocities u and a range of shear-rates $\dot{\gamma}$ are shown in Figure 9b. The B10 columns
422 (i.e., the bimodal columns with the smallest contributions of fine particles) appear to segregate
423 the slowest, however, we found no clear relationship between t_{seg} and any parameters. Comparing
424 Figure 9b with Figure 8, it is evident that the segregation time is greater than the time required
425 for the rheometer to collect the first shear stress measurements, but it is significantly shorter than
426 the total duration of the experiment.

427
428 The analysis of the shear stress data shows that an aerated bimodal granular mixture subjected
429 to shear does not have the same shear stress at the beginning and at the end of the measurement.
430 This implies that the particle distribution and hence the flow pattern of the mixture both change
431 in time whilst undergoing aeration and shearing processes. This in turn highlights the importance
432 of considering the applied shear when predicting the state of mixing or segregation of the mixture
433 for designing systems relying on the different behaviour of the solid phases.

434 4. Conclusions

435 This paper reports an experimental investigation into the effects of shear on particle segrega-
436 tion of aerated bimodal granular columns. The columns were composed of Ballotini glass beads of
437 nominal diameters $63\text{ }\mu\text{m}$ and $355\text{ }\mu\text{m}$ mixed in four different proportions. The granular columns
438 made up of 50%, 40%, 25%, and 10% $63\text{ }\mu\text{m}$ by mass were compared to the results of two monodis-
439 perse columns, comprised 100% $355\text{ }\mu\text{m}$ and 100% $63\text{ }\mu\text{m}$, respectively. The granular mixtures were

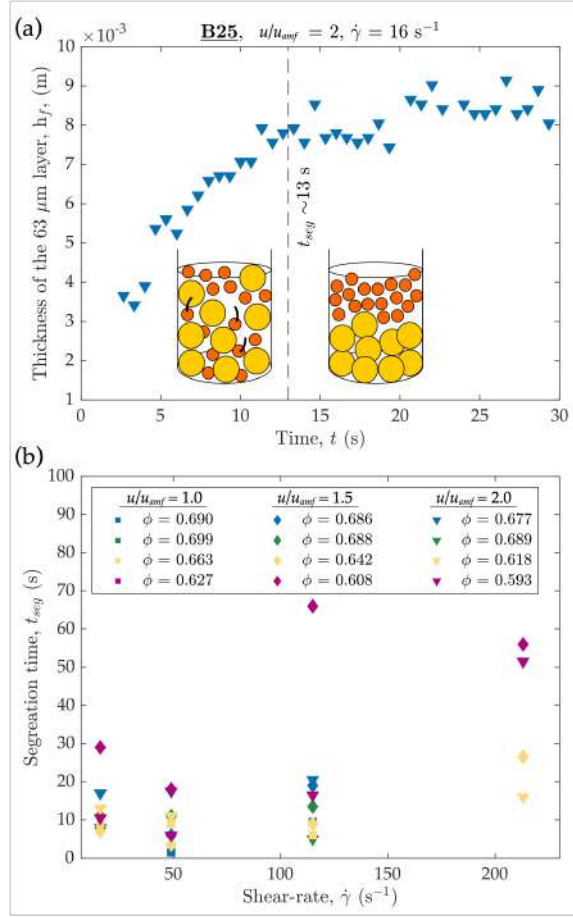


Figure 9: (a) Thickness of the upper fine layer, h_f , as a function of time, t for a bimodal mixture B25 fluidised at air velocity $u = m \text{ s}^{-1}$ and sheared at $\dot{\gamma} = 16 \text{ s}^{-1}$. The dashed line represents the segregation time, t_{seg} . (b) The segregation time, t_{seg} , for all the segregated bimodal granular columns for a range of air velocities, u and shear rates, $\dot{\gamma}$.

440 sheared at shear rates of $8 \text{ s}^{-1} \leq \dot{\gamma} \leq 213 \text{ s}^{-1}$ while simultaneously fluidised by a flux of compressed
441 air at velocities $0 < u \leq 2 u_{mf}$, where u_{mf} is the apparent minimum fluidisation velocity.
442 Regardless of the shear-rate applied, $\dot{\gamma}$, all the granular columns expand when aerated with air
443 velocities $u > 0 \text{ m s}^{-1}$. At sufficiently high air velocities, air bubbles form at the base of the column
444 and move vertically upwards towards the column surface. For monodisperse columns, the column
445 exhibits bubbling fluidisation, while, for the bimodal columns, the $63 \mu\text{m}$ glass beads move towards
446 the top of the column and the column segregates into a layer of $355 \mu\text{m}$ glass beads overlain by a
447 layer of $63 \mu\text{m}$ particles. In bimodal columns, the applied shear was observed to suppress bubble
448 formation, resulting in reduced particle segregation. Additionally, the results show that the seg-
449regation of particles affects the shear stress and hence the flow patterns of the granular mixture,
450 therefore highlighting the the importance of considering the applied shear when predicting the state
451 of mixing, mobility, or segregation of granular mixtures.

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456 Data availability statement

457 Data supporting the findings of this study are openly available at [10.5281/zenodo.14651636](https://doi.org/10.5281/zenodo.14651636).

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