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Dynamical behaviour of fine granular glass/bronze mixtures under vertical vibration

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Abstract It has recently been reported that mixtures of fine equal-sized bronze and glass spheres separate into glass-rich and bronze-rich regions under sinusoidal vertical vibration [Burtally et al., Science 295, 1877 (2002)]. Here we report a study of these separation effects, varying both the bronze/glass size ratio and the composition of the mixture. For a wide range of mixtures we observe separation into bronze-rich and almost pure glass regions separated by extremely sharp boundaries. Either a bronze-rich region forms above a glass layer or a bronze-rich layer is sandwiched between two glass layers. Convection exists within each layer but does not act to cause mixing of the two regions. The separation into two regions is maintained even in the presence of a variety of spatial oscillations. The variations of behaviour with differing glass/bronze size ratios support the proposal of a differential air-damping separation mechanism, while experiments using a porous bottomed box indicate that separation requires air to be driven through the granular bed by the vibration.

1 Introduction

The dynamical behaviour of a granular material, under vertical vibration, is well known to be extremely non-linear and to be strongly influenced by the inelastic nature of the collisions between grains [1] and the collisions between the grains and the container walls. Vertical sinusoidal vibration of sufficient amplitude induces convection, surface waves and arching [2–7]. For shallow layers of grains, pattern formation and oscillons are observed [8,9]. The ratio of the peak acceleration of the container to that of gravity, $\Gamma = a\omega^2/g$, is an important parameter governing the motion. Here a is the amplitude of vibration, ω is the angular frequency and g is the acceleration due to gravity. The onset of convection generally occurs for Γ somewhat exceeding unity, while patterning, surface waves and arching require appreciably higher values of Γ [2–9].

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For fine particulate systems, air effects are also important and introduce a wealth of new phenomena. If a single component granular bed is vibrated vertically upon a horizontal surface, the presence of air induces piling and convection [10]. Fine grains, vibrated vertically within a container, break symmetry by forming a tilted upper surface down which grains cascade [11,12]. Faraday suggested that grains are sucked under the main body by the moving air as the supporting platform accelerates downwards. Later, the granular body falls upon these grains, causing continuously erupting piles [10]. Thomas and Squires [13] have offered a related explanation in which grains are carried towards the center of the bed by the downward and inward air currents that occur during bed flight. However, alternative explanations of the Faraday effect have been offered and it is the subject of active debate [13–16].

If a single larger and heavier grain is introduced into a vertically vibrated granular bed, the intruder tends to move to the upper surface, the “Brazil nut effect” [17]. A larger and lighter intruder may, under appropriate circumstances, move to the bottom, the “reverse Brazil nut effect” [18]. Computer simulations also predict conditions for the “Brazil nut” to take up an intermediate height [19]. Recently, the presence of air has been observed to modify the behaviour of the intruder [20].

There is also a substantial body of knowledge on the separation and segregation of granular mixtures [17,21, 22,24,25]. Recent simulations [26], and analytic theories [27] have offered criteria for separation based upon mass and size differences. However, to date, many of these theoretical predictions have not been tested experimentally. Furthermore, much of this work has concentrated on mixtures of large particulates for which air effects are expected to be unimportant.

Recently Burtally et al. have reported that fine mixtures of equal sized glass and bronze spheres separate under vertical vibration into glass-rich and bronze-rich regions with extremely sharp interface boundaries [28]. At lower frequencies the bronze-rich region is observed to form an upper layer above an almost pure glass region, while at higher frequencies the bronze-rich layer forms as a sandwich between upper and lower glass regions. In both cases, the motion of the bronze spheres is notably more active than that of the glass and the separation phenomena disappear at low air pressures. Although a mechanism based on the differential damping effect of the air upon the two species has been proposed to explain these phenomena, a detailed understanding is still lacking.

Here, we go beyond this first brief report, describing a detailed series of experiments on vertically vibrated fine bronze-glass mixtures. First, we describe the principle features of the separation phenomenon and the dynamic behaviours found following separation. Secondly, we report the effects of varying the bronze/glass composition. Thirdly, we vary the size ratio of the two components in experiments designed to probe the air-damping mechanism. Finally, we then describe experiments conducted using a porous bottomed box. These show that air is necessary for separation to occur, but that the presence of air is not in itself sufficient. Rather the air must be forced through the grains during vibration. These experimental findings lead to a more detailed explanation of the basic separation mechanism and many of the observed effects.

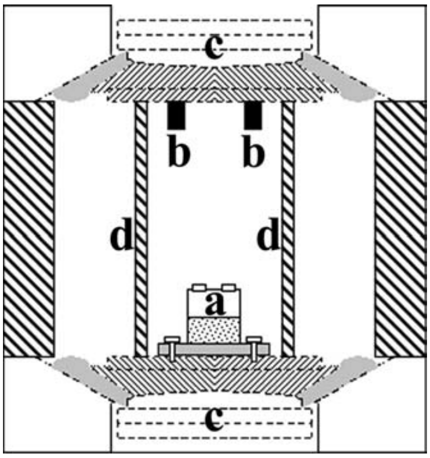

Fig. 1. View of the apparatus showing the glass box and box mount (a), the accelerometers (b), the double transducer assembly (c) and the connecting frame (d)

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Experimental details

The experiments which we shall describe use bronze spheres of density $\rho_b = 8900\,\mathrm{kg/m^3}$ and soda-glass spheres of density $\rho_g = 2500\,\mathrm{kg/m^3}$. The bronze was obtained from Makin Powders Ltd or sourced from Eckart-Werke GbH. The glass was obtained from Potters Ballotini Ltd.

Spheres of closely similar sizes often form close packed crystalline structures under vibration, with bulk movement of a whole crystalline block rather than independent movement of individual grains. The glass and bronze spheres were therefore sieved to produce a size-spread in order to avoid crystallisation effects. Each sample was taken from a source having a very wide size spread. Consequently, the size distribution is close to uniform over each of the narrow ranges quoted below.

We have studied four ratios of the mean diameters. Mixtures **A** consist of bronze spheres with diameters in the range $125-150\,\mu$m and glass spheres with diameters in the range $63-90\,\mu$m. Mixtures **B** consist of bronze in the range $90-125\,\mu$m and glass in the range $90-125\,\mu$m. Mixtures **C** consist of bronze in the range $63-90\,\mu$m and glass in the range $125-150\,\mu$m, while mixtures **D** consist of bronze in the range $45-53\,\mu$m and glass in the range $125-150\,\mu$m. For each class of mixture, we have studied volume ratios of the bronze to glass components of 25% : 75% (referred to as 1), a 50% : 50% mix (referred to as 2) and a 75% : 25% mix (referred to as 3).

A chosen mixture, of mean depth 20 mm, was contained within a rectangular soda-glass box 50 mm high and of internal dimensions $40\,\mathrm{mm} \times 10\,\mathrm{mm}$ in the horizontal plane. The boxes were vibrated using the arrangement shown in Fig. 1. The glass box was glued to a metal mount, which was then bolted to a rigid frame held between a pair of electromagnetic transducers. The transducer assembly was attached to a massive concrete block (~ 250 kg). This configuration, with its extreme stiffness against transverse motion, ensured accurate one dimensional motion over the frequency range of interest ($10\,\mathrm{Hz} < f < 200\,\mathrm{Hz}$), the axis of vibration being aligned to the vertical to within 0.2^o. The vertical motion was monitored with a pair of capacitance cantilever accelerometers, covering the ranges $0-8\,g$, and $0-80\,g$. The waveform of the vibration was monitored on an oscilloscope to check the accuracy of its sinusoidal nature. The motion of the box is distorted from sinusoidal by less than 0.5% of the amplitude through the frequency range 20 Hz to 200 Hz.

Persistently shaken fine glass spheres develop static charge. If vigorous shaking is continued for some time grains may stick to the walls, impairing photography. Importantly, the accumulation of static limits the maximum time over which measurements may be made on a particular mixture, the build up of cohesion slowing or even halting the progress of some of the phenomena which we will describe. The accumulation of static may be made worse by working in a vacuum or by eliminating moisture through baking. The addition of minute quantities of an anti-static surfactant may somewhat slow the build up of static without appreciably influencing the dynamics. However in the present studies, we have preferred to replace the mixture being studied with a freshly prepared one should the cohesive effects of static become appreciable. Our experiments are conducted in a laboratory with a controlled humidity lying in the range 35−45%.

While we observe no difference in the behaviour of mixtures made using the alternative bronze supplies, differences are found in the behaviour of mixtures formed from different production batches of glass spheres. These differences lie in the exact conditions for the separation phenomena to occur, rather than in the principal phenomena themselves. There is some correlation between these behavioural differences and the maximum angle of stability of the glass, θ_m, determined by very slowly tipping a freshly formed flat surface until avalanching is observed. For the materials we have considered, θ_m ranges from 35^o to 43^o. Those batches with lower θ_m, are also observed to be more free flowing and to build up charge less rapidly upon prolonged vibration. The θ_m of a granular material principally depends upon the shape of the grains, their surface friction and any cohesion due to excessive moisture. In all batches used in the present work the majority of the grains are observed to be close to spherical, while baking tests show that moisture is not a contributing factor to the magnitude of θ_m. It is therefore likely that the observed variation of θ_m is due to variations of surface friction. Those batches with higher friction might be expected to build up charge more rapidly during vibration, as is indeed observed. In this paper we have chosen to initially report the behaviour of mixtures formed from batches of glass with higher values of θ_m. At the end of each section we will describe the differences in behaviour when a more free flowing batch of glass, having lower θ_m, is used.

In order to categorise the different separation behaviours we consider mixtures subject to different regimes of f and Γ and display the boundaries between different behaviours in the f, Γ plane. In these figures the visual judgement used to distinguish between some types of behaviour is only based on a semi-quantitative assessment. In addition, there are some sample to sample variations in behaviour. However, for all like mixtures we observe the same general features of separation and dynamic behaviour, and the same topology of the boundaries within the f, Γ plane, the principal interest of this paper. Such plots are therefore a useful means of summarising the range of observed behaviours.

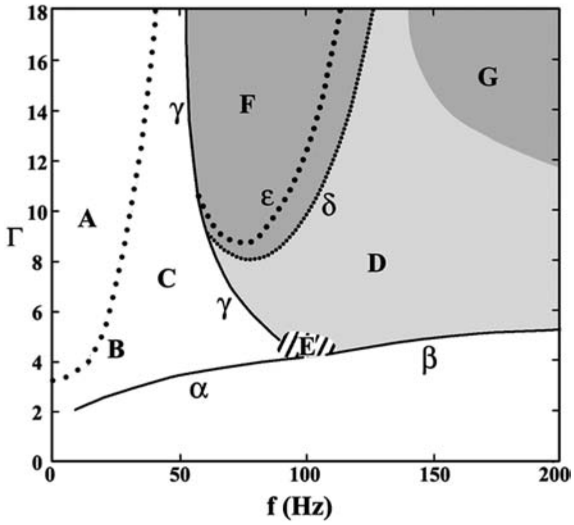

Fig. 2. Schematic behaviour of mixture **A**1, as a function of frequency and Γ, showing the onset of bronze-on-top (α), the onset of sandwich separation (β), the transition boundary between the two (γ), and the onset of slow (δ) and rapid (ϵ) asymmetric inversion oscillations. Also shown are the areas of bronze-on-top (A) and (C), granular throwing at the upper surface, and upper surface and interface fluctuations (A), simple tilt oscillations (B), sandwich formation (D), oscillations between the bronze-on-top and sandwich configurations (E), continuous asymmetric inversion oscillations (F) and continuous symmetric inversion oscillations (G)

3
The separation phenomena

An outline of the behaviours observed for a mixture **A**1 at various values of f and Γ is shown schematically in Fig. 2. The boundaries to the various behaviours and features shown have been obtained principally by applying particular values of f and Γ to a granular sample in the well mixed state, and observing the subsequent behaviour. These same boundaries have also been probed by slowly increasing or decreasing either f or Γ, keeping the other variable constant.

At lower frequencies convection and Faraday tilting of the upper surface are observed as the amplitude of vibration is increased. Once convection begins there is a tendency to form bronze-rich and glass-rich regions with diffuse boundaries. Separation proceeds very slowly. At higher values of Γ separation proceeds more quickly and sharp separation boundaries appear between the glass-rich and bronze-rich regions. The bronze-rich regions rapidly merge into a single bronze-rich region which lies above a lower glass-rich region. Fig. 3 illustrates this form of separation for 35 Hz and $\Gamma = 5.6$. The figure shows that the development of separation occurs by a process of coarsening, an effect observed in other granular systems [29]. Both regions become close to homogeneous throughout their individual volumes as the process of separation continues. At values of f and Γ corresponding to the line α of Fig. 2, separation into the two regions with "bronze-on-top" is substantially complete in two minutes. The eighth image (centre bottom) of Fig. 3 corresponds to an example of what we judge to be "substantially complete" separation. For lower values of Γ the formation takes far longer. However, the separation process may take a shorter time if the initial mixture is substantially inhomogeneous.

Following separation, the boundary between the two regions is extremely well defined, being only one grain-diameter wide. Once bronze-on-top separation is complete, convection currents occur within the individual bronze and glass-rich regions but they do not act to cause mixing.

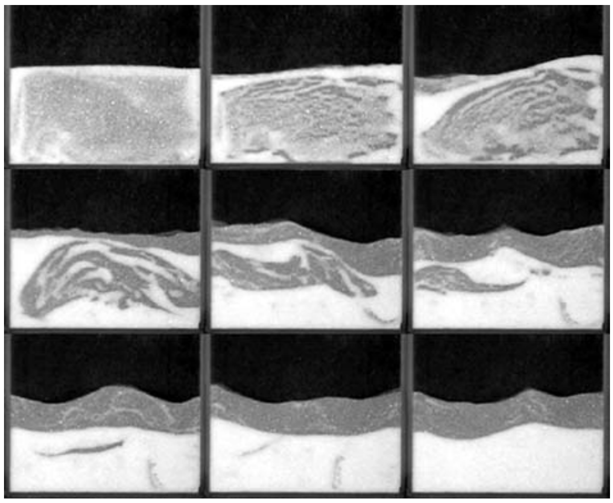

Fig. 3. Behaviour of mixture **A**1 under $\Gamma = 5.6$ and 35 Hz showing the formation of the sandwich configuration. The pictures form a time sequence from upper left to lower right and correspond to 0, 0.5, 1, 2, 3, 6, 30, 60 and 70 seconds of vibration

After prolonged vibration at fixed f and Γ the bronze-rich region eventually contains a small proportion of glass, estimated by visual inspection following the cessation of vibration, to lie in the range from 20% to below 1% depending upon f and Γ. The lower region consists almost entirely of glass.

Above α, in the bronze-on-top (unshaded) areas A and C of Fig. 2, the spatial form of both the separation boundary and the upper surface fluctuate in time. At low values of f and Γ, in the area around B, simple oscillations back and forth between the two alternative tilts occur [30]. At higher values of f and Γ, the fluctuations contain both periodic and non-periodic components. The area A indicates approximately the values of f and Γ for which bronze grains are visibly thrown free of the upper surface. In this area considerable fluctuations of both the interface and the upper surface are observed. Surprisingly, a sharp separation boundary between the distinct regions is maintained despite these disturbances. However, in the higher parts of A the fluctuations and throwing from the upper surface become so severe that the maintenance of separation into two distinct regions fails following a particularly severe contortion. Glass from the lower glass region breaks through the upper bronze-rich layer, mixing with the bronze.

At higher frequencies there is also a line, β in Fig. 2, at which sharp separation boundaries between a bronze-rich region and glass-rich regions appear on a time scale of two minutes. Separation may be achieved on a longer time scale at somewhat lower values of Γ. Here, however, the bronze-rich regions rapidly merge to form a stable single layer at an intermediate height, between upper and lower glass-rich regions. This we refer to as the "sandwich" configuration. Again, the bronze-rich region contains a proportion of glass but the glass-rich regions are almost completely free of bronze. The formation of a sandwich configuration is illustrated in Fig. 4. In the early stages the central region contains pockets rich in glass but these are then ejected as the sandwich develops. In the sandwich areas, D, F and G, shown shaded in Fig. 2, the glass regions are eventually almost pure while the bronze-rich layer generally contains 4−20% of glass depending upon f and Γ.

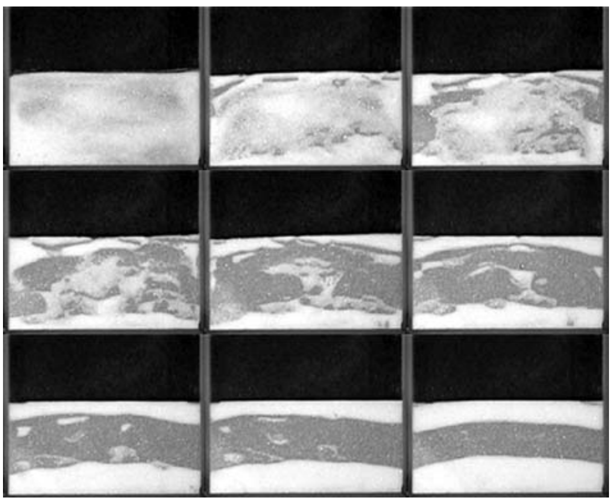

Fig. 4. Behaviour of mixture **A**1 under $\Gamma = 6.8$ and 160 Hz showing the formation of the sandwich configuration. The pictures form a time sequence from upper left to lower right and correspond to 0, 1, 2, 3, 4, 11, 60, 120, and 300 seconds of vibration

In the sandwich area, D, and for some higher values of Γ, the formation of glass droplets may be observed within the otherwise homogeneous bronze-rich region. These drops then either fall into the lower glass layer or rise to join the upper glass layer. It appears that glass is continually passing into the bronze-rich layer and that this process maintains equilibrium.

In the area E of Fig. 2, where lines α and β meet, the development of separation involves complex oscillations between the bronze-on-top configuration of area C and the sandwich configuration of area D.

In either separated configuration, convection may be observed within the individual glass and bronze-rich regions, with considerable velocity shear at the sharp separation boundaries, yet no convection currents are present which would mix the regions. In areas A and C this shear is particularly evident since, in the tilted configurations found there, the bronze-rich and glass-rich regions generally have rapid convection currents of opposite direction. Both at low and at high frequencies, we observe considerably more kinetic activity in the bronze-rich region than in the glass-rich region with correspondingly greater speeds of convection in the former.

The boundary between the bronze-on-top area, C, and the shaded sandwich zone is shown as the line γ in Fig. 2. As this line is approached from below, or from the left, the upper bronze region avalanches down the tilted slope and passes into the bulk as a cylindrical roll, then spreading out to form a sandwich between upper and lower glass-rich layers. We refer to this process as an asymmetric inversion. At no stage are the sharp separation boundaries between the bronze-rich and glass-rich regions lost. If the line γ is approached from the right the sandwich configuration slowly changes to the bronze-on-top configuration, the configuration transforms continuously by the glass diffusing from the upper glass layer through the bronze-rich region into the lower glass layer. At lower values of Γ this can take a very long time indeed; movements of the order of 1mm take periods of order 1 hour. We have been unable to observe for longer times due to the build up of static effects which inhibit the motion.

In the area indicated as F in Fig. 2 an asymmetric inversion process repeats continuously with some fluctuations in the time-period. One half period of such a process is shown in Fig. 5. Upon entering F from area D the bronze-rich layer of the sandwich rises slowly to the surface, while remaining close to horizontal. Once there, the surface tilts. The bronze then avalanches down the slope, and passes into the bulk of the glass to form a stable low-lying horizontal layer, reforming the sandwich. The bronze then slowly rises to the surface again and the process is repeated. At the line marked δ the mean period of this repeating process is about five minutes, while by the line ϵ the mean period has fallen to about one minute.

Within the area G an alternative type of oscillation is found, a form of symmetric inversion. Such a process is shown in Fig. 6. The bronze layer moves towards the upper surface of the bed, but before it reaches the upper surface it quite suddenly necks symmetrically and divides in the middle, glass flowing rapidly upwards through the gap. The two bronze volumes then pass down the right and left sides of the box to reform a single sandwich layer low

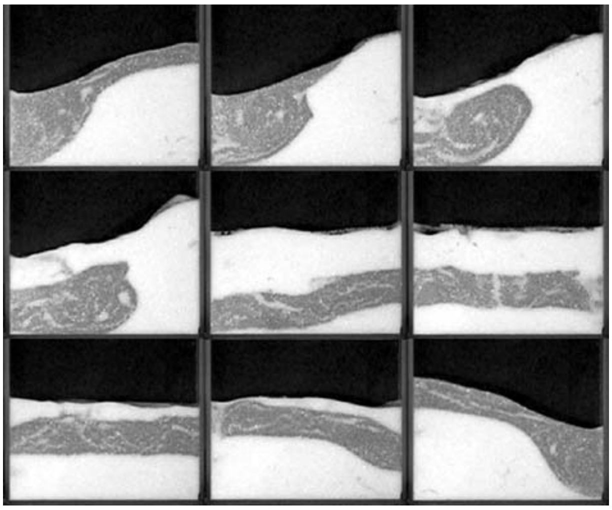

Fig. 5. Behaviour of mixture **A**1 under $\Gamma = 16.7$ at 70 Hz showing one half of an asymmetric inversion oscillation. The pictures form a time sequence from upper left to lower right. The total period of the oscillation is about 7 s

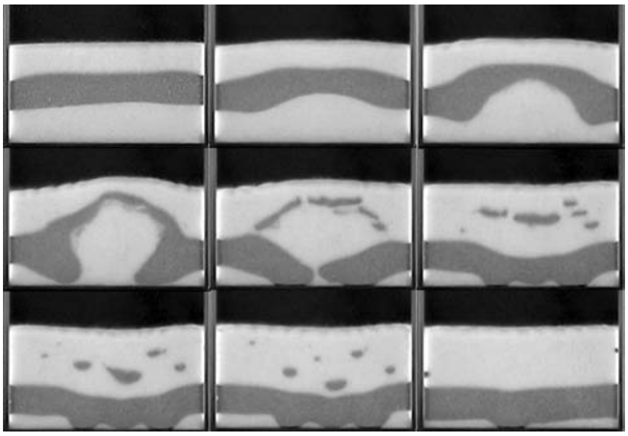

Fig. 6. Behaviour of mixture **A**1 under $\Gamma = 16$ at 170 Hz showing one period of a symmetric inversion oscillation. The pictures form a time sequence from upper left to lower right. Note the formation of a number of small bronze fragments, which stay intact, later joining the main body of bronze. The total period of oscillation is about 37 s

in the bed. During this process the main bodies of bronze shed a number of small fragments. Each of these sharp-boundaried fragments remains intact, eventually joining the main bronze-rich sandwich layer. This single horizontal bronze-rich layer then slowly moves towards the surface and the process repeats. The onset of this form of oscillation appears after variable amounts of time and the boundary to this form of behaviour, shown in Fig. 2 as G, is only approximate.

In almost all of the experiments within the 10 mm × 40 mm box the separation is close to two dimensional, the configuration of the bronze-rich and glass-rich regions being very similar when viewed through the opposite large faces of the box. One exception to this is the spatial contortion of the interface and upper boundaries of the bronze-on-top configuration found at low frequencies and higher values of Γ. Another is for the small bronze fragments formed during inversion processes, which are three-dimensional objects.

If these experiments are repeated on a mixture **A**1 formed using more free flowing glass, bronze-on-top separation is again observed at low frequencies and sandwich separation is observed at high frequencies. However, the onset of granular convection and the positions of the separation lines α and β occur at lower values of Γ. The lines α and β occur at values of Γ lower by about 20% at 150 Hz but only 5% lower at 50 Hz. While the boundary line, γ, between the bronze-on-top and sandwich configurations is of the same form as in Fig. 2 it is displaced somewhat to higher frequencies and the area E occurs at about 125 Hz rather than 100 Hz. In the high frequency regime the region of inversion oscillations in which the bronze rises to the top and then inverts to reform the sandwich configuration now extends far lower in Γ. The build up of static is far slower for mixtures using the more free flowing glass and it has been possible to extend our investigations to oscillation periods approaching an hour. Even so, time restrictions make it impossible to determine how close to the β line long period inversion oscillations extend. In these mixtures the inversion oscillations cannot be categorised into the asymmetric or symmetric kind since a range of asymmetry is observed between the symmetric form found at higher frequencies to the asymmetric form found just above the γ boundary line.

4
Varying the composition

In this section we will study the effect of varying the bronze/glass composition by comparing and contrasting the behaviour of mixtures **A**1, **A**2 and **A**3, where the bronze and glass diameters are 125–150 μm and 63–90 μm and the bronze percentages by volume are 25%, 50% and 75% respectively. An outline of the behaviour of mixture **A**2 as a function of f and Γ is shown schematically in Fig. 7. As with mixture **A**1, at low frequencies we observe tilting and the onset of separation into a bronze-on-top configuration, the line marked α indicating substantially complete separation into the bronze-rich and glass-rich regions in 2 min. At higher frequencies we observe separation into the sandwich configuration, the 2 min onset occurring at the line β. At the intersection of the α and β lines there is a small area marked E within which complicated oscillations between the bronze-on-top and the sandwich configurations occur.

We note in Fig. 7 the area A in which visible throwing of grains from the surface occurs together with vigorous fluctuations of the interface and upper surface. However, for this mixture composition and $\Gamma < 18$, there is no failure of the bronze-on-top separation despite this activity. Simple tilt oscillations occur in the area around B. The boundary between the extensive bronze-on-top area, C, and the shaded sandwich area, D, is also shown. For values of Γ progressively further above the β line, the top layer of the sandwich reduces in thickness. If Γ is steadily increased the top layer may be observed to thin by diffusion of glass through the central bronze layer. At sufficient values of Γ the upper layer fails to cover the bronze

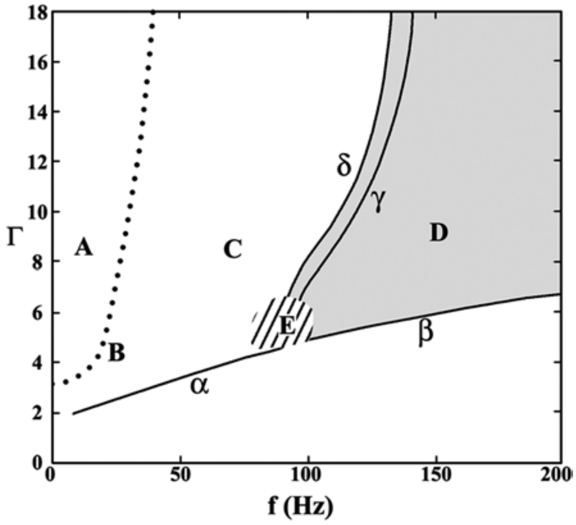

Fig. 7. Schematic behaviour of mixture **A**2, as a function of frequency and Γ, showing the onset of bronze-on-top (α), the onset of sandwich separation (β), the transition between incomplete-sandwich and bronze-on-top (δ), and the transition between incomplete and full sandwich (γ). Also shown are the areas of bronze-on-top (A) and (C), area of granular throwing at the upper surface, and upper surface and interface fluctuations(A), simple tilt oscillations (B), sandwich formation (D) and area of oscillations between the bronze-on-top and sandwich configurations (E)

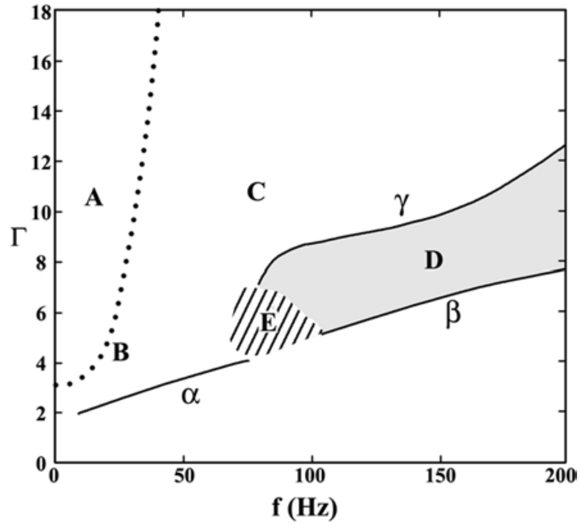

Fig. 8. Schematic behaviour of mixture **A**3, as a function of frequency and Γ, showing the onset of bronze-on-top (α), the onset of incomplete-sandwich separation (β) and the transition boundary between the two (γ). Also shown are the regions of bronze-on-top (A) and (C), granular throwing at the upper surface, and upper surface and interface fluctuations (A), simple tilt oscillations (B), and incomplete sandwich formation (D). Oscillations between the bronze-on-top and the incomplete-sandwich configurations occur within E

layer completely, by the line γ. We refer to this configuration as an "incomplete-sandwich". For sufficiently high Γ the upper glass is completely absent and the bronze-on-top configuration is found, at the line γ. When the upper surface is symmetrically domed due to twin convection cells, an arrangement often found at higher frequencies, the incomplete-sandwich has glass both at the right and left-hand extremes of the box. Incomplete-sandwiches also occur when the upper surface is tilted. Glass may then be found only at either the upper or the lower regions of the slope. For mixture **A**2 the tilted configuration is found close to region E while at higher frequencies the symmetrical configuration is preferred.

For mixture **A**2 we find no inversion oscillations. If Γ is kept constant, while f is varied to take the system between areas C and D, the process of transformation between the bronze-on-top and sandwich configurations is very slow and the behaviour is hysteretic even on time scales of many minutes.

The equivalent information for mixture **A**3 is shown schematically in Fig. 8. Once again, we observe the formation of bronze-on-top separation within two minutes at the line α. For **A**3 however we observe no full sandwich formation, rather the onset of a region of symmetric incomplete-sandwich formation at the line β. Oscillations between the two configurations occur within the intermediary area E. As Γ is increased above the line β, the upper glass regions of the incomplete-sandwich retreat, the full bronze-on-top configuration being recovered by the line γ. We observe no inversion processes anywhere in the f, Γ plane, transformations of configuration occurring by the slow diffusion of glass through the bronze-rich layer.

When the experiments are repeated with an **A**2 mixture containing more freely flowing glass the α and β lines are found to be somewhat lower in Γ. When values of f and Γ are applied at higher frequencies, a sandwich forms within an area similar to D of Fig. 7. However the configuration then changes by diffusion to the bronze-on-top form on a time scale of tens of minutes to over one hour depending upon f and Γ.

If the experiments are repeated with an **A**3 mixture made using more free flowing glass, the α line is similarly found to be lower in Γ than that of Fig. 8. If values of f and Γ are applied, corresponding to D of Fig. 8, an incomplete-sandwich forms. However, it lasts only about the same time as that taken for separation itself to occur. The separation passes quickly to the bronze-on-top configuration. The β line is now effectively an extension of the α line, the onset of the bronze-on-top configuration, which for this mixture is the stable form throughout the upper part of the diagram.

In comparing the behaviours of **A**1, **A**2 and **A**3, we note the following. All mixtures exhibit excellent separation with extremely sharp separation boundaries. The onset of separation with increasing Γ occurs at lines α and β which rise only slightly as the bronze proportion is increased. The region of bronze-on-top behaviour increases with increasing bronze in the case of the mixtures using less free flowing glass. The region of sandwich behaviour found in **A**1 withdraws to a restricted area of incomplete-sandwich behaviour by **A**3. For mixtures with free flowing glass, the bronze-on-top form soon dominates throughout as the bronze proportion increases.

If similar experiments are repeated in boxes having a more square cross-section very similar processes of separation into bronze-on-top, sandwich or incomplete-sandwich configuration occur. However, inversion operations are effectively suppressed by the use of a 10 mm × 10 mm box. Restricting the box dimension in this way also suppresses the tilt oscillations in the area around B.

5 Varying the size ratio

Burtally et al. [28] investigated equal sized bronze/glass mixtures and suggested that the separation process may be based on the difference in the effect of the air damping on the two components. If this is the case, the strength of the separation and the spatial arrangement of the separated configuration will depend upon the size ratio of the two components, as well as their densities. We now report experiments carried out to investigate this size dependence.

As a first approximation, the influence of the air on the bronze and glass components may be estimated by treating each bronze and glass sphere as interacting with air in a manner described by Stokes' law. For a mixture of particles of densities ρ_1 and ρ_2 and mean diameters d_1 and d_2, the relative strength of the air-effects on the two components may then be characterised by the ratio $S = (\rho_1 d_1^2)/(\rho_2 d_2^2)$. Here, S is just the ratio of the accelerations of the two components induced by the same fluid flow. This approximation ignores spatial variations in the air velocity and some corrections to Stokes' law are required, particularly at the velocities found at higher Γ. Nevertheless, as we will see, S is a useful parameter in comparing the behaviours of four types of mixture having different size ratios.

For the mixtures **A**1, **A**2 and **A**3 which we have just discussed $S = (\rho_b d_b^2)/(\rho_g d_g^2) \approx 12$, the glass component being far more heavily damped than the bronze. These mixtures display separation with sharp interface boundaries and with clear regions of bronze-on-top, and sandwich behaviour. The separation boundaries, once formed, are stable against considerable disturbance.

The mixtures **B** have $S \approx 3.6$, the glass component being slightly more damped than the bronze. Preliminary data for a mixture of composition **B**1 has already been reported by Burtally et al. [28]. Figure 9 shows schematically the results of a more extensive study, using the same time-scale criteria as above. The similarities to the data for **A**1 should be noted. The separation lines, α, for bronze-on-top and β for sandwich formation are again evident, as is the line γ at which transformation between the two configurations occurs. This transformation again takes place by inversion when approaching from below or from the left. The area F of Fig. 9 is one of asymmetric inversion oscillations. An area of symmetric inversion oscillations occurs within the region G.

The behaviour of mixture **B**2 has many similarities to that of **A**2. Sandwich behaviour is now found over a limited range of Γ at frequencies above 95 Hz. The transformation into bronze-on-top as Γ is raised occurs by diffusion, passing from a sandwich through an incomplete-sandwich configuration. In **B**3 only incomplete-sandwich behaviour is observed over a limited range of Γ values and above 150 Hz.

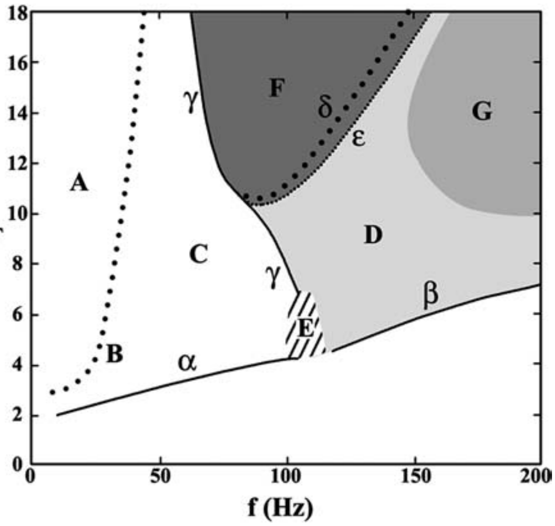

Fig. 9. Schematic behaviour of mixture **B**1, as a function of frequency and Γ, showing the onset of bronze-on-top (α), the onset of sandwich separation (β) the transition boundary between the two (γ), and the onset of slow (δ) and rapid (ϵ) asymmetric inversion oscillations. Also shown are the regions of bronze-on-top (A) and (C), area of granular throwing at the upper surface, and upper surface and interface thrashing (A), simple tilt oscillations (B), sandwich formation (D), oscillations between the bronze-on-top and sandwich configurations (E) and continuous asymmetric inversion oscillations (F). In the area G continuous symmetric inversion oscillations occur

If **B** mixtures are formed using glass from a more free flowing batch the differences in behaviour are very similar to those for the A mixtures. The α and β lines are somewhat lower in Γ, the inversion oscillations of **B**1 extend to lower values of Γ and the bronze-on-top configuration becomes more dominant in mixtures **B**2 and **B**3.

The principal differences between the behaviours of the **A** and **B** series of mixtures are as follows: the bronze-rich regions of the **A** mixtures contain more glass than the **B** mixtures do under corresponding conditions; the formation of glass droplets within the bronze-rich region is far more evident for mixtures **A** than for **B**; the dynamics of the **A** series are appreciably faster than for the **B** series over much of the f, Γ plane.

For the mixtures **C** the parameter $S \approx 1$. For **C**1 we observe very poor separation, with traces of sharp boundaries only visible over the limited frequency range of 25−90 Hz at lower values of Γ. The formation of bronze-enhanced regions close to the upper surface are observed at lower frequencies and bronze-enhanced regions at intermediate levels at some higher frequencies. We also observe slow oscillatory behaviours of the bronze-enhanced regions. However, the separation is always very poor; the sharp separation boundary, where it does exist, distinguishes regions containing a considerable proportion of the other component. At all frequencies increasing Γ readily induces global convection currents which thwart any tendency to separate. Mixtures **C**2 and **C**3 exhibit even weaker tendencies to separate.

For the mixtures **D**, $S \approx 0.4$. The air-damping of the bronze is now greater than that of the glass. For mixture **D**1, separation with glass uppermost is experienced at all frequencies investigated as Γ is slowly increased, as is shown in Fig. 10, where the shaded area corresponds to the "glass-on-top" configuration. For this area the separation boundaries are sharp. Above the shaded area strong global convection acts to thwart attempts to separate. Local areas of sharp boundary may be seen and attempts at sandwich formation may be identified at higher frequencies. Nevertheless separation into bronze-rich and glass rich regions is always far from complete, separation always competing with mixing convection. As the bronze concentration is increased the band of Γ values for which "glass-on-top" separation is found narrows and for **D**3 the sharpness of the boundary between the glass-rich and the bronze-rich regions is more diffuse.

If **D** mixtures are formed from more free flowing glass, a predominance of glass occurs at the upper surface once granular motion is established by increasing Γ. However the formation of separate glass rich and bronze rich layers is thwarted by the nature of the convection which is global rather than established in two separate regions. Glass continually passes to the upper surface where it predominates. However, glass is also continually passing by convection down the side walls of the container into the bronze-rich lower region.

In summary, we observe a very strong separation mechanism for mixtures **A**, particularly for those richer in glass. Small bronze-rich regions remain intact with sharp boundaries even when separated from the main bronze body under vigorous dynamics. Mixtures **B** exhibit a tendency to separate which is only slightly weaker than mixtures **A**. For mixtures **A** and **B** the dominant configuration at low frequencies is for the bronze to separate to the top. For mixtures **C** where the two species are estimated to be equally damped only a very weak separation is exhibited, with a slight tendency for bronze to be uppermost at low frequencies. For mixtures **D** there is a tendency to separate with glass at the top. This is however appreciably weaker than for **A** and **B** and is thwarted at higher Γ, or for more free-flowing mixtures, by global convection.

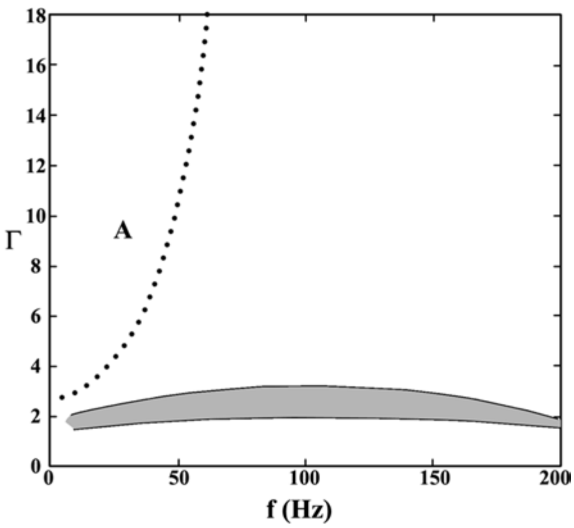

Fig. 10. Schematic behaviour of mixture **D**1, as a function of frequency and Γ, showing the area of area of granular throwing at the upper surface, and upper surface and interface fluctuations (A), and the "glass on top" configuration shown shaded. At all values of Γ above this shaded area, global convection competes with the tendency to separate

Thus, our mixtures with $S >> 1$ separate well with bronze-on-top as the low frequency configuration. The mixture with $S \approx 1$ separates only weakly with evidence for the bronze-on-top configuration over a range of low frequencies. The mixtures with $S < 1$ show a weak tendency to separate with the glass component uppermost. Between $S \approx 1$ and $S \approx 0.4$ the separation order has clearly reversed. The crude basis for the use of the parameter S as the ratio of the influence of air damping on the two species has been noted. Nevertheless, these results provide strong support for separation mechanism being based upon differential air-damping.

6
Further experiments

Burtally et al. have reported that the separation of mixture **B**1 disappears at sufficiently low pressures which vary approximately linearly with frequency. The pressures reported are in reasonable agreement with a theory based on treating the system as a porous medium and using Darcy's law to describe the air flow [28]. We have repeated these experiments on mixtures **A** and **B**. Vibration is applied to cause separation and the pressure is slowly lowered. In the low frequency regime the separation boundaries become more diffuse as the pressure is lowered, while the rate of convection increases, with appreciable downward convection at the larger faces of the box. Convective currents eventually cause global mixing. In the high frequency regime the separation fails by the separation boundaries becoming more diffuse as the pressure is lowered and the bronze-rich layer broadening as it absorbs more and more glass. We find similar pressures for the failure of separation for the two mixtures and can further confirm the approximate linear dependence of these pressures with frequency.

We have further tested the involvement of air in the separation process by conducting experiments on a $10\,\text{mm} \times 40\,\text{mm}$ box constructed with glass walls but with a porous bottom. The bottom surface consists of a layer of $63\,\mu$m woven steel sieve mesh supported by a 3 mm layer of metal foam, itself supported periodically by pillars. This structure is extremely porous to air, while being rigid to particle collisions. The top of the box may be open or closed.

With the top of the box open, we find no tendency to separate in any of our mixtures. Rather we observe global mixing convection with one or more clear convection cells. In this configuration the air is free to move during the vibration. With the top of the box closed, however, the air is forced through the bed during the motion. We then observe the clear separation of mixtures **A** and **B** with sharp separation boundaries and with the bronze-rich region uppermost. It is clear that the presence of air is a necessary but not sufficient condition for separation to occur. The air must be forced through the granular bed by the vibration, as it is for a solid-based container.

7 Discussion

We have produced evidence for very strong separation effects both in glass/bronze mixtures of equal size but also in other mixtures, particularly those where the parameter S is considerably greater than unity. These separation effects are noteworthy, both for the very sharp interfaces between homogeneous bronze-rich and glass-rich regions and for the immunity of these regions and their interfaces to violent disturbances. We have shown, for example, that both large and small fragments of the bronze-rich region may circulate during inversion oscillations, with the interface intact. A curious but key feature of the separation is the occurrence of convection solely within each component, rather than global convection that would cause mixing.

For each of the mixtures **A** and **B** we can find conditions which produce separation in less than a minute in which the glass is essentially pure and the bronze contains of order 1% glass by volume.

It is clear that the separation mechanism is based on the interstitial air. Separation of this type disappears for large particles [28]. The separation effects also vanish at low pressures and the values and frequency dependence of the failure pressure support an air-based hypothesis. Furthermore, if the mechanism was not air based, the separation found in the porous bottomed box should not be influenced by the top being closed.

The supposition that it is the differential air damping of the two species is supported by the present experiments in which the parameter S is varied. By varying S we can change both the strength and the order of separation.

What of the strong separation mechanism itself? Our present experiments using a porous bottomed box have shown that just the presence of air is not sufficient to cause separation. If air is free to move up and down with the granular bed, no separation occurs. However, in a solid bottomed box, air is forced through the bed as it moves with respect to the box during the vibration. Strong separation may then occur. The case of the porous bottomed box with a sealed top is intermediate between these two situations since, due to the volume of air above the granular bed, air is only partially forced through the bed during vibration. Bronze-on-top, but not sandwich formation, is then observed.

It is clear from these key pieces of evidence that the separation mechanism is more subtle than that proposed by Burtally et al. [28]. It is necessary for grains of the same species to be acted upon by approximately the same force, that induced by the air being driven through the bed. The two species respond differently to this driven air, because of their different mass and/or diameters. Consequently, grains of the same species which have come together will then subsequently tend to move together since they experience the same force due to the air. This process will repeat on larger and larger length scales, causing the system to coarsen until only one or two regions of a particular species are present.

In the present system, the relative velocities of adjacent grains are usually small compared to the maximum velocity of the container. Under these circumstances, gaps between the different separated regions may occur over an appreciable fraction of the vibratory cycle. Indeed, under spectroscopic illumination of appropriate phase, gaps are clearly visible between the glass and bronze regions of the separated configurations. These gap stabilise the separation by enabling convection to occur within the individual regions; relative convective motion occurs when the regions are not actually in contact. The resulting convection patterns do not then act to cause mixing.

Given a strong tendency for the glass and bronze to separate, many of the features we observe can be readily explained. The more damped region, the glass-rich region in the case of mixtures **A** and **B**, will be relatively compact and inert. It will be difficult for bronze to re-enter such a region. The bronze-rich region on the other hand will be relatively dilate; glass will find it possible to enter. Equilibrium will be maintained by the formation of glass droplets and by their subsequent movement. In mixture **A** the bronze is larger than in mixture **B**; there will be more interstitial space in the bronze rich region. This would explain why we observe a greater proportion of glass within the bronze-rich regions of **A** than within those of **B** with correspondingly greater droplet formation in **A**.

Confirmation of the separation mechanism and study of the dynamics may be provided by suitable computer simulations. Preliminary simulations, based on the use of Stokes' law to incorporate the viscous damping, confirm that separation is indeed dependent upon air being forced through the bed [31].

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