Gravitational instabilities in binary granular materials

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The motion and mixing of granular media are observed in several contexts in nature, often displaying striking similarities to liquids. Granular dynamics occur in geological phenomena and also enable technologies ranging from pharmaceuticals production to carbon capture. Here, we report the discovery of a family of gravitational instabilities in granular particle mixtures subject to vertical vibration and upward gas flow, including a Rayleigh–Taylor (RT)-like instability in which lighter grains rise through heavier grains in the form of “fingers” and “granular bubbles.” We demonstrate that this RT-like instability arises due to a competition between upward drag force increased locally by gas channeling and downward contact forces, and thus the physical mechanism is entirely different from that found in liquids. This gas channeling mechanism also generates other gravitational instabilities: the rise of a granular bubble which leaves a trail of particles behind it and the cascading branching of a descending granular droplet. These instabilities suggest opportunities for patterning within granular mixtures.

granular material | instabilities | fluidization

From moving sand dunes (1) to avalanches (2), granular motion forms the basis of several natural phenomena. These dynamics are also utilized in pharmaceuticals production (3) and carbon capture (4). Previous studies have found fascinating similarities between granular and liquid flow, e.g., lizards swimming through sand (5), wavy instabilities found in grains under shear (6), gas bubbles rising through ensembles of particles (7, 8), Faraday waves in granular layers (9, 10), and particle droplets forming in hourglass streams (11) and tapped plates (12). The Rayleigh–Taylor (RT) instability (13) is a classic fluid mechanical phenomenon in which lighter fluid pushes into denser fluid above it due to buoyancy, producing “fingers” and “bubbles” of lighter fluid. This instability manifests itself in a broad range of systems, from the formation of mushroom-shaped clouds in nuclear explosions (14) to fingering patterns in supernova explosions (14) to structures in lava lamps (15). A variety of fingering patterns (16–18) have been observed in grains, including density-driven, RT-like phenomena when a closed cell containing liquid (19) or gas (20) and particles is quickly inverted and the fluid pushes upward on the falling particles.

The understanding and prediction of RT behavior between two types of grains is of great significance in multiple scientific fields, including geological structures ranging from mud-sand sediments underwater (21) to deposits under glaciers (22) to volcanic flows (23). Two differences from traditional fluid flow prevent RT behavior from occurring between dry grains: (i) grains under pressure have a solid-like nature with strong frictional forces preventing them from flowing like a fluid and (ii) grains typically do not have significant cohesive forces to create a surface tension between two types of granular material, and surface tension provides the stabilizing force in the classic understanding of the RT instability. Previous studies have shown that seismic shaking of granular material saturated with water can cause liquefaction, enabling grains to move and create RT-like structures (24). “Fluidization” of granular material based on the upward flow of gas or liquid suspending the particles can also allow grains to flow like liquid. However, prior studies have shown

that upward flow of liquid leads to either a homogeneous “layer inversion” (25) if both particles are fluidized, or the formation of volcano-like “water escape structures” (26) if only the lighter grains are fluidized. Fluidization via gas flow creates gas bubbles which rise through the system, causing the formation of granular structures very distinct from those associated with the RT instability (27). Vibration of dry grains with interstitial gas causes buoyant segregation (28), but without net gas flow, interstitial gas does not lead to the liquefaction necessary for RT structures to form. Here, we seek to address the question if an analog of the RT instability exists between dry grains of different density when exciting a granular system using a combination of vibration and gas flow and, if so, how the underlying mechanism compares with that occurring in conventional fluids.

Our experiments consist of a model binary granular system with two sets of spherical particles of different diameters and densities (color-coded) arranged in a classic RT setup (Fig. 1 and Movie S1) with the light particles (black) below the heavy particles (white). The cell, a “vibrated-fluidized bed,” shakes vertically to induce motion in the grains and contains upward airflow at velocities close to the “minimum fluidization velocity,” U_{mf} , needed to suspend the particles by providing a drag force which offsets their weight. Under these conditions, both sets of particles exhibit liquid-like behavior, i.e., they are loosely packed and can flow. However, importantly, the system does not exhibit any gas bubbling or bulk convection of particles, as would happen if the gas flow or vibration strength were increased. The U_{mf} of both sets of particles are similar because the light particles have a larger diameter than the heavy particles ($U_{mf} \propto d^{1/2} \rho_p$), where d is the particle diameter and ρ_p is the particle density.

Fig. 1 reveals that when light particles are placed below heavy particles in the system and vibration and gas flow are initiated, an RT-like instability is observed, with “granular bubbles” of less-dense

Significance

Flow of granular materials, such as sand and catalytic particles, is critical to a wide range of natural phenomena and industrial processes. However, the physics underlying granular flows is poorly understood. We report the discovery of gravitational instabilities in binary granular materials driven by a gas channeling mechanism unseen in fluids. A Rayleigh–Taylor instability analog is observed despite a lack of surface tension between grains, while a droplet branching phenomenon demonstrates a deviation from fluid motion due to solidification of particles. These discoveries shed light on granular dynamics and can potentially explain geological formations and enable process technologies.

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Fig. 1. Time series of optical images of the RT instability forming when denser particles are placed initially above lighter particles. Diameter of light particles: $d_L = 1.70 \pm 0.07$ mm; density of light particles: $\rho_L = 2,500$ kg/m³; ratio of heavy-to-light particle diameters and densities: $d_H/d_L = 0.69 \pm 0.03$, $\rho_H/\rho_L = 2.40$; Geldart (42) grouping of both types of particles: Group D; vibration frequency: $\omega = 188$ rad/s; vibration strength: $\Gamma = A\omega^2/g = 0.18$, where A is the amplitude of the vibration; height, width, and depth of the cell: $H/d_L = 235$; $W/d_L = 118$; $D/d_L = 4.7$; gas: humid air at ambient conditions, density: $\rho_g = 1.19$ kg/m³, viscosity: $\eta_g = 1.8$ Pa s; minimum fluidization velocity needed to suspend the particles at these vibration conditions for light and heavy particles: $U_{mf,L} = 0.84$ m/s, $U_{mf,H}/U_{mf,L} = 1.03$; superficial gas velocity: $U/U_{mf,L} = 1.17$.

particles pinching off from fingers and rising to the surface, until a complete inversion and segregation of particles is reached. We define granular bubbles as locally concentrated ensembles of light particles surrounded by heavy particles through which the granular

bubbles rise. Fig. 2 shows that this RT-like instability is not limited to the conditions in Fig. 1, but rather persists across different particle sizes, vibration conditions, and ratios of minimum fluidization velocity for the heavy and light particles ($U_{mf,H}/U_{mf,L}$). Fig. 2 further

Fig. 2. Structures formed for RT experiments under different flow conditions: (A) varying gas flow rate, (B) varying particle size, (C) varying vibration strength, (D) varying ratio of minimum fluidization velocities of the two particles ($U_{mf,H}/U_{mf,L}$). All images have the same cell dimensions, gas properties, particle densities, and Geldart (42) grouping of both types of particles as in Fig. 1. Each panel of images varies a property while keeping other properties constant: (A) constant properties across images: $d_L = 1.70 \pm 0.07$ mm, $d_H/d_L = 0.69 \pm 0.03$, $\omega = 188$ rad/s, $\Gamma = 0.18$, $U_{mf,H}/U_{mf,L} = 1.03$; $U/U_{mf,L}$ varies between images: (A, i) 0.88, (A, ii) 0.94, (A, iii) 1.00, (A, iv) 1.05, (A, v) 1.11, (A, vi) 1.17, (A, vii) 1.35. (B) Constant properties across images: $\omega = 188$ rad/s, $\Gamma = 0.18$, $U/U_{mf,L} = 1.17$; particle size varies: (B, i) $d_L = 0.61 \pm 0.06$ mm, $d_H/d_L = 0.67 \pm 0.06$, $U_{mf,H}/U_{mf,L} = 1.10$ (B, ii) $d_L = 1.70 \pm 0.07$ mm, $d_H/d_L = 0.69 \pm 0.03$, $U_{mf,H}/U_{mf,L} = 1.03$. (C) Constant properties across images: $d_L = 1.70 \pm 0.07$ mm, $d_H/d_L = 0.69 \pm 0.03$, $\omega = 188$ rad/s; vibration strength varies: (C, i) $\Gamma = 0.09$, $U/U_{mf,L} = 1.14$, $U_{mf,H}/U_{mf,L} = 0.97$, (C, ii) $\Gamma = 0.18$, $U/U_{mf,L} = 1.17$, $U_{mf,H}/U_{mf,L} = 1.03$ (C, iii) $\Gamma = 0.36$, $U/U_{mf,L} = 1.18$, $U_{mf,H}/U_{mf,L} = 1.10$. (D) Constant properties across images: $\omega = 188$ rad/s, $\Gamma = 0.18$, $U/U_{mf,L} = 1.17$; varying properties between images: (D, i) $d_L = 1.64 \pm 0.05$ mm, $d_H/d_L = 0.71 \pm 0.03$, $U_{mf,H}/U_{mf,L} = 1.10$, (D, ii) $d_L = 1.70 \pm 0.07$ mm, $d_H/d_L = 0.69 \pm 0.03$, $U_{mf,H}/U_{mf,L} = 1.03$, (D, iii) $d_L = 1.70 \pm 0.07$ mm, $d_H/d_L = 0.64 \pm 0.03$, $U_{mf,H}/U_{mf,L} = 0.98$. (E) Shows a regime map for gas bubbling occurring, granular fingering occurring without gas bubbles and no interfacial motion as a function of vibration strength and the ratio of superficial velocity to $U_{mf,L}$ of the light particles without vibration.

Fig. 3. Numerical simulation results near the interface for the RT configuration. Each dot corresponds to an individual particle. The system size is the same as that used experimentally in Fig. 1, but the images zoom in on the interface to provide more detail. Rows show progression over time. The first column shows the positions of particle types. The second column shows the vertical component of gas velocity normalized by the inlet gas velocity. The third column shows the net buoyant force determined by the sum of the vertical component of drag ($F_{d,z}$) and gravitational ($F_{g,z} = -w_p$) forces normalized by particle weight (w_p). The fourth column shows the sum of frictional and normal contact forces ($\Sigma F_{c,z}$) in the vertical direction on each particle normalized by the particle weight. Diameter of light particles: $d_L = 1.70$ mm; density of light particles: $\rho_L = 2,500$ kg/m³; ratio of heavy-to-light particle diameters and densities: $d_H/d_L = 0.65$, $\rho_H/\rho_L = 2.40$; Geldart (42) grouping of both sets of particles: Group D; vibration frequency: $\omega = 63$ rad/s; vibration strength: $\Gamma = 0.20$; height, width, and depth of the cell: $H/d_L = 235$; $W/d_L = 118$; $D/d_L = 5.8$; gas density: $\rho_g = 1.2$ kg/m³, gas viscosity: $\eta_g = 1.8$ Pa s; ratio of minimum fluidization velocities: $U_{mf,H}/U_{mf,L} = 1.10$; superficial gas velocity: $U/U_{mf,L} = 1.25$.

shows that the size and growth rate of the fingers increases with increasing gas flow rate, vibration strength, and particle size. Experiments with only gas flow or only vibration were unable to

produce the phenomenon (*SI Appendix, Fig. S3*), and Fig. 2*E* shows that the window of gas velocities which enables fingering without gas bubbling increases with increasing vibration strength.

Fig. 4. Optical images of instabilities in suspended granular flows in three configurations: (A) a granular bubble of lighter particles below a sea of denser particles, (B) a homogeneous mix of denser and lighter particles, and (C) a droplet of denser particles above a sea of lighter particles. Other than the initial configuration of particles, the conditions are identical to those in Fig. 1.

Fig. 5. Granular bubble rise velocity (U_b) versus (A) the effective bubble diameter ($D_{b,eff}$), which is calculated using $D_{b,eff} = 2L_b\pi^{-1/2}$, where L_b is the side length of the initial square of light particles at $U/U_{mf,L} = 1.03$ and (B) normalized superficial gas velocity ($U/U_{mf,L}$) with $D_{b,eff} = 34$ mm. For both A and B, the same particles and vibration conditions as in Fig. 4 were used. Error bars show the SD from three repetitions of each experiment. The lines show least-square regressions through the data: the red dotted line is a linear fit, and the black solid line is a square-root fit.

The conventional RT instability involves buoyancy acting as a driving force for inversion and surface tension acting as a stabilizing force for the interface, resulting in the formation of fingers of a characteristic wavelength or width. The mechanism driving the formation of these structures in fluidized grains is perplexing because the grains are suspended by gas flow and have no significant cohesive forces to create surface tension. The phenomenon can be explained as follows: Buoyancy drives the inversion, since the two particle–gas mixtures have different bulk densities. This difference in bulk density has been used to describe a homogeneous “layer inversion” in particles suspended by upward liquid flow (25) as well as wavy deformation structures in liquid-saturated sediments subject to vibration (24). Initially, a high effective “viscosity” of the grains acts to stabilize the interface, causing the formation of perturbations of a characteristic wavelength instead of a homogeneous inversion. Particle-phase viscosity arises due to interparticle friction and has been used to explain finger formation when granular particles sediment in a liquid (29), a process which also lacks surface tension. The wavelength producing the maximal finger growth rate predicted by viscous potential flow theory for viscous fluids with no surface tension (29) is given by $\lambda = 4\pi(\eta_H + \eta_L)^{2/3}g^{-1/3}(\rho_{B,H}^2 - \rho_{B,L}^2)^{-1/3}$, where g is the acceleration due to gravity. Assuming a viscosity of $\eta \sim 1$ Pa s for both sets of fluidized particles (30) and using the bulk densities of both particle/air mixtures ($\rho_{B,H}$, $\rho_{B,L}$) gives a wavelength of $\lambda \sim 40$ mm, which is in line with the width of perturbations which develop into fingers in Fig. 1. Thus, the competition between buoyant and viscous forces describes the initial formation of fingers in the model binary granular system. The ratio of gas flow rate to the minimum fluidization velocity (U/U_{mf}) is also key to the formation of these perturbations because gas flow is needed to suspend the particles, enabling them to act like a mobile viscous fluid, rather than a stationary solid. It is worth noting, however, that the equation given earlier for wavelength does not depend directly on the gas flow

rate. The regime map in Fig. 2E shows that vibration strength is another key dimensionless group for characterizing this phenomenon because vibration is needed to lower the gas velocity at which the particles become suspended, allowing fingers to form without the presence of gas bubbles. *SI Appendix, Fig. S4* shows that U/U_{mf} and the normalized vibration strength Γ are the appropriate non-dimensional parameters for describing the excitation mechanisms necessary for the RT-like phenomenon, since these two parameters collapse data for the fingering regime map across different particle sizes and vibration conditions.

While these mechanisms and nondimensional groups properly describe the initial perturbation of the interface, they do not fully describe the behavior of fingers after the initial formation stage. The fact that the description is incomplete is demonstrated by the trend that finger width increases with gas velocity, as shown in Fig. 24. The effective viscosity of the suspended particles decreases with increasing gas flow rate due to a decrease in the number of interparticle contacts. Thus, the wavelength equation given earlier predicts a decrease in wavelength with increasing gas velocity, yet developed fingers are wider at higher gas velocities.

To uncover the missing physics underlying this RT-like phenomenon after the initial perturbation stage, we simulate a system with the same size and particle properties as in the experiments in Fig. 1 using a numerical model which couples gas and particle motion (31). Fig. 3 shows that there is a competition between a net buoyant force pushing light particles at the interface up, while a net contact force pushes them down. Due to the gas flow and vibrations, small-amplitude waves are able to form at the interface, and the width of these perturbations which grow into fingers is ~ 40 mm, consistent with those observed experimentally. Upon formation of these wavy perturbations, the drag force acting on particles at the crest of a wave increases dramatically, causing the wave to grow into a finger and ultimately pinch off as a granular bubble. This increase in drag force occurs due to gas channeling through the light particles (second column of Fig. 3) because they are larger than the heavy particles, and thus have a higher permeability to gas flow (32), $k \propto d^3$. Such gas channeling has been used to explain vertical band structures forming in binary gas-fluidized systems with different particle size, but of the same particle density (27). With an increasing gas flow rate, more gas can channel through wider waves, allowing wider fingers to form and grow, explaining the trend seen in Fig. 24 and distinguishing the instability from that seen in conventional fluids. The ratio of the vertical component of the gas velocity through fingers and surrounding heavy particles will be equal to the ratio of gas permeability between the two types of grains $u_{z,L}/u_{z,H} = k_L/k_H$ (27), which scales with the ratio of particle diameters $k_L/k_H \propto (d_L/d_H)^3$. Importantly, when the same simulation was run, but with interparticle friction turned off, large gas bubbles appeared which caused particle motion which was very different from the RT-like instability, demonstrating the importance of interparticle friction and thus effective viscosity on the formation of granular fingers.

Fig. 4 shows how differences in bulk density and permeability between light and heavy particles lead to a variety of other interesting instabilities under the same gas flow and vibration

Table 1. Rise velocity of granular bubbles with different particles

Light particle diameter (d_L), mm	Diameter ratio, d_H/d_L	Density ratio, ρ_H/ρ_L	Granular bubble rise velocity, mm/s
0.61 ± 0.06	0.67 ± 0.06	2.40	13.0 ± 2.7
1.70 ± 0.07	0.69 ± 0.03	2.40	42.9 ± 1.9
2.75 ± 0.13	0.69 ± 0.04	2.40	11.0 ± 3.4
1.70 ± 0.07	0.77 ± 0.04	1.60	10.5 ± 1.1

Vibration frequency of 188 rad/s, vibration strength of $\Gamma = 0.18$, $U/U_{mf,L} = 1.13$ under these vibration conditions and a light particle density of $\rho_l = 2,500 \text{ kg/m}^3$.

Fig. 6. Particle image velocimetry images of the velocity field (colors indicate speed and arrows indicate velocity) of light particles surrounding heavy particles under the same fluidization and vibration conditions as in Fig. 4C. The white region surrounded by black lines demarcates the “granular droplet” of heavy particles.

conditions. When a granular bubble of light particles is surrounded by a sea of heavy particles (Fig. 4A), gas channeling through the light particles causes them to rise together as a bubble to the top, leaving behind a trail of particles due to the lack of surface tension. Fig. 5A shows that the rise velocity increases with granular bubble diameter, but it does not follow the correlation $U_b \propto D_b^{1/2}$ observed in classic fluids and gas bubbles in fluidized grains (33). Additionally, the rise velocity also increases monotonically with superficial gas velocity (Fig. 5B), further emphasizing the deviation from traditional fluid flow. This monotonic increase deviates from the nonmonotonic change in rise velocity of an intruder particle with increasing gas velocity in a bed of vibrated granular material (34). Since the RT-like phenomenon and the granular bubble rise phenomenon have the same underlying mechanisms, the trends and governing parameters for granular bubble rise are expected to be the same between both phenomena. Table 1 demonstrates that the rise velocity of a granular bubble increases with increasing density ratio (ρ_H/ρ_L) and decreasing diameter ratio (d_H/d_L). This result affirms that increased buoyant driving force and increased gas channeling due to increased permeability ratio (k_L/k_H) lead to increased upwelling rates of granular bubbles and granular fingers. Varying particle size has a nonmonotonic effect on granular bubble rise velocity, demonstrating the complexity of the phenomenon. It is worth noting that the granular bubble shown in Fig. 4A initially accelerates before reaching a constant rise velocity; thus, the rise velocities shown in Fig. 5 and Table 1 are based on the constant rise velocity reached after the initial acceleration, as shown in *SI Appendix, Fig. S2*.

When the heavy and light particles are initially mixed (Fig. 4B), light particles ultimately reach the top due to buoyancy. During segregation, finger- and granular-bubble-like structures form because gas channels through regions with a higher concentration of light particles, causing these regions to rise and further increase in concentration of light particles as they rise.

When a “droplet” of heavy particles is surrounded by a sea of light particles (Fig. 4C) the droplet does not fall straight to the bottom as in immiscible fluids, but rather it immediately splits into two daughter droplets which fall on an angle through the light particles and undergo subsequent binary-splitting events. The observed binary splitting of granular droplets exhibits striking similarity to the bifurcations of falling dye droplets in miscible fluids (35–39). However, the physical mechanisms behind these two phenomena are different. When a droplet of dye

is placed in a miscible fluid, it forms a vortex ring. This ring becomes unstable and fragments into smaller droplets which undergo the same instability and may fragment again (37). The RT instability is suspected to cause the fragmentation of the vortex ring (38, 39). The mechanism underlying the granular binary splitting reported here can be explained as follows: Gas channels around the granular droplet because of the lower permeability of the heavy particles. The lack of gas flow to suspend the light particles just below the granular droplet causes a region below the droplet to solidify, or become stationary, due to force chains under the weight of the droplet. The solidification of particles below the droplet is confirmed experimentally using particle image velocimetry (40) in Fig. 6. The droplet cannot fall through these solidified particles, but it still can descend through the suspended particles, and thus it splits and falls at an angle through the suspended light particles just above the solidified particles. At a certain point, a solidified region forms just below the daughter droplets for the same reason of gas channeling, and thus they undergo a subsequent splitting event. Thus, the mechanism of gas channeling and solidification of particles makes this branching phenomenon distinct from that observed in conventional miscible fluids.

Thus, the combination of gas channeling and differences in bulk density provides a mechanistic explanation of the physics underlying heterogeneous flow behavior observed in the discovered gravitational instabilities in suspended binary granular flows reported here. Due to similarities and differences in governing physics, the resulting patterns sometimes mimic those observed in conventional fluids, but have different underlying mechanisms.

Materials and Methods

The vibrated-fluidized beds were constructed using polymethyl methacrylate and vibrated using an electrodynamic shaker (ET-139; Labworks Inc.). The airflow rate was controlled using a mass-flow controller (F-203AV; Bronkhorst) and passed through a humidifier to keep a constant relative humidity of 87–91%. “Light” particles were spherical glass beads, and “heavy” particles were spherical ceramic beads; particles were purchased from Sigmund Lindner GmbH, and full particle properties are given in *SI Appendix, Table S1*. Minimum fluidization velocities were measured using the pressure drop across the bed of particles as the gas velocity was slowly decreased (*SI Appendix, Fig. S1*). Optical images were obtained using a CCD camera (Photron Fastcam SA Z), and image processing was used to determine the rise velocity of granular bubbles and the particle velocity field surrounding granular droplets; full details are given in *SI Appendix*. Simulations were conducted

using the Computational Fluid Dynamics – Discrete Element Method (CFD-DEM) (31) using CFDEMcoupling software (41).

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