Origin of Stability in Sedimentation

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Velocity fluctuations and particle concentrations are studied in a liquid fluidized bed to investigate the origin of steady state sedimentation. Both the velocity fluctuations and the particle concentrations are found to strongly depend on height. A flux balance model shows why the bed is stable: velocity fluctuations drive a downward particle flux that is compensated by an upward particle flux stemming from an excess flow velocity due to the stratification in concentration. Our results show that in steady state the magnitudes of the fluctuations are related to the degree of stratification.

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The slow sedimentation of a collection of monodisperse spheres in liquids is a fundamental problem in physics and is of wide importance in chemical reactors [1]. In the "classical" picture, sedimentation proceeds in a stable and uniform way; macroscopic quantities such as the average concentration and sedimentation velocity do not vary with position or time during settling. Recent experiments, however, have shown large nonuniformities on the microscopic, or particle, scale [2–6]. Local concentrations and velocities were found to fluctuate substantially in time about their mean values. The implications of the fluctuations on the macroscopic properties have not been fully addressed, in particular, whether the widely used [1–7] classical picture of macroscopic uniformity represents a stable, steady state configuration in the presence of such large fluctuations. On this latter point, Tee *et al.* [8] recently found that initially uniform suspensions can destabilize and become highly stratified in concentration as the particles settle. Most importantly, the system never stabilized; the stratification in concentration continuously evolved over the entire settling time [8]. These observations suggest that the classical picture of macroscopic homogeneity is not valid, but leave unanswered the important question of how, or indeed whether, steady state sedimentation occurs.

To search for a steady state, the subject of this Letter, one would naturally like to allow the system as much time as possible to evolve. In this regard, sedimentation experiments are inherently limited, eventually all of the particles are on the bottom of the cell. Much longer times, however, can be achieved using a fluidized bed, where liquid is pumped upwards through the particles to create a drag force that counteracts gravity. If properly balanced, the average particle velocities in the lab frame are zero, and the particles are perpetually sedimenting.

In this Letter, we describe several experimental findings in a liquid fluidized bed. First, there exists a condition of steady state sedimentation, evidenced by near zero time averaged velocities v_x and v_z, and local volume fractions ϕ, rms velocity fluctuation magnitudes σ_v, and velocity correlation lengths ξ that are constant in time. Second, ϕ, σ_v, and ξ are strongly height dependent. Third, the local sedimentation velocity v_{sed}, measured just after turning off the upward flow, is also strongly height dependent and is smaller than the velocity v_{pump} at which fluid is pumped upwards to stabilize the particle column. The excess velocity is defined as $v_{\text{ex}}(z) = v_{\text{pump}} - v_{\text{sed}}(z)$. We use the continuity equation to develop a criterion for stability and explicitly show that the particle column is stable because at all positions the particle flux gradient *downwards* due to fluctuations in ϕ and v is nearly equal and opposite to the flux gradient *upwards* due to the stratification in the product ϕv_{ex}. The balancing of flux gradients yields a stability condition $\partial(\phi v_{\text{ex}})/\partial z = -1.1\sigma_v\sqrt{S(\phi)\phi a^3/\xi^5}$ that shows that the magnitudes of the fluctuations are related to the stratification in concentration and excess velocity.

We use spherical glass beads of radius $a = 137 \pm 13\ \mu$m dispersed in a mixture of glycerol and water with a viscosity of $\eta \approx 70$ cp. The Stokes settling velocity of an isolated sphere is measured and determined to be $v_{\text{St}} = 0.63 \pm 0.01$ mm/s. Particle motions are at high Peclet number (Pe $\approx 10^9$), low Reynolds number (Re $\approx 10^{-3}$), so that Brownian diffusion and inertial forces are negligible. The temperature is at the ambient value $T = 21$ °C. The sample cell is a rectangular glass tube of dimensions $D \times W \times H = 8$ mm $\times$ 80 mm $\times$ 305 mm. The bottom of the cell is glued into a metal base into which the water/glycerol mixture is continuously pumped. The overflow liquid at the top of the cell recirculates back into the pump, forming a closed loop. To enable a uniform flow into the cell, a silica filter is glued across the entrance to the cell at the bottom. With the pump off, the spheres form a sediment ~2.1 cm tall, containing ~800 000 particles. When the pump is on, the particles expand upward, filling a region above the bottom up to a height dependent upon the pumped fluid velocity v_{pump}. We set $v_{\text{pump}} = 0.525$ mm/s, which expands the particle column to a height $h_{\text{bed}} \sim 18.5$ cm, with an average volume fraction $\langle\phi_{\text{bed}}\rangle = 0.638(2.1/18.5) \sim 0.072$. It takes approximately 1000 s, or 3 times the settling time $\tau_h = h_{\text{bed}}/v_{\text{pump}} \sim 350$ s, for the particle column to reach a steady state.

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Stable particle columns are observed for more than 5 h, or $> 80\,000$ Stokes times $\tau_{St}(= a/v_{St})$.

Particle velocities are measured using the technique of particle image velocimetry (PIV) [9]. The apparatus consists of a (1008×1016 pixels) CCD camera, a synchronized stroboscope illuminating the cell from behind, and specialized image processing hardware and software from Dantec Instruments. The depth of field of the camera lens is ≈ 5 mm. A large cross section of the cell is imaged (3.4 cm $\times$ 3.4 cm) so that several thousand particles can be simultaneously studied. Velocity maps consisting of 62 vectors $\times$ 62 vectors are extracted by comparing two closely timed pictures using standard PIV techniques. Each vector is the average velocity of 2 to 4 spheres. Local particle volume fractions are determined from the local optical turbidity. An expanded laser beam passes through the fluidized bed onto a CCD camera that records the transmitted laser intensity I_T. Volume fractions ϕ are determined by comparing I_T with a calibration curve for $I_T(\phi)$ measured at the mid height of particle columns of mean concentration $0.02 \leq \langle\phi_{bed}\rangle \leq 0.15$.

Figure 1 shows typical velocity vector maps and photographs from a stable fluidized bed, where (a) corresponds to a position near the top and (b) to a position near the middle of the particle column. For scale, we also show the magnitude of the fluid velocity v_{pump} pumped upwards through the bed. Both velocity maps show regions moving both upwards and downwards, but the magnitudes of the velocities are significantly larger near the middle than near the top. Moreover, the photographs reveal that the particle concentration is fairly uniform in the middle of the particle column, but markedly less uniform as well as less concentrated near the top. To quantify these observations, we measure the velocity maps and the local volume fractions at different heights along the particle column. We extract from the velocity maps the mean velocities, $v_x = \langle v_{i,x}\rangle$, and $v_z = \langle v_{i,z}\rangle$, and the root mean square (rms) velocity fluctuations, $\sigma_v = \langle v_z^2\rangle^{1/2}$, where $\langle\ldots\rangle$ represents an average over $\sim$50 vector maps of 3844 vectors each. As seen in Fig. 2, the average velocities in the horizontal and vertical directions, v_x and v_z, are close to zero over the entire height of the bed, which defines the stable steady state.

The strong height dependence of the fluctuation magnitude σ_v is also in evidence in Fig. 2. Close to the bottom there is substantial mixing, $\sigma_v/v_{pump} \sim 0.8$, while near to the top, $\sigma_v/v_{pump} \sim 0$.

To determine the characteristic length scale of the velocity fluctuations, we fit the correlation function $C_z(z) = \langle v_z(0)v_z(z)\rangle/\langle v_z(0)^2\rangle$ to the form $C_z(z) = \exp(-z/\xi)$, with results for ξ shown in the inset of Fig. 2. The correlation lengths are not uniform in height and exhibit a decrease towards the top part of the column.

Moreover, the local volume fraction, ϕ, shows a dependence on height as well. Figure 3 shows that ϕ varies from $\phi \sim 0.10$ at the bottom to $\phi \sim 0.035$ at the top, with the mean concentration of the column, $\langle\phi_{bed}\rangle = 0.072$, found near to the middle. Note also the sharp interface at the top, with ϕ dropping from $\phi \sim 0.035$ to $\phi \sim 0.00$ within $\sim$0.5 cm.

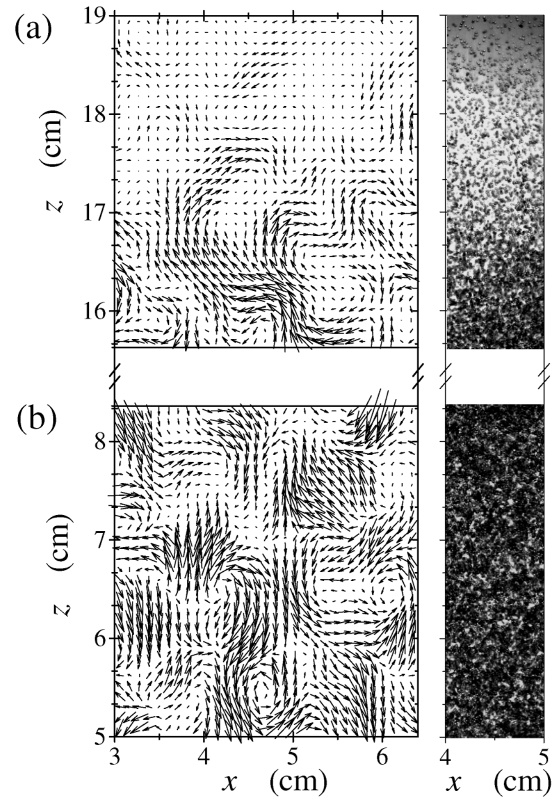

FIG. 1. Velocity vector maps and photographs of a stable fluidized bed at $\langle\phi_{bed}\rangle = 0.072$. Figures 1(a) and 1(b) correspond to respective positions near to the top and near to the middle of the particle column. The single arrow on the right gives the corresponding scale of the fluid velocity, $v_{pump} = 0.525$ mm/s, at which fluid is pumped upwards through the bed to stabilize it. Note that the velocity scale in (a) is magnified relative to (b) by a factor of 3 for clarity.

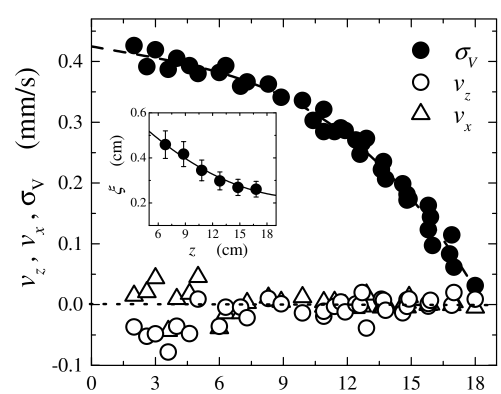

FIG. 2. Average velocities v_x and v_z and velocity fluctuations σ_v as a function of height z in a stable fluidized bed. Inset: correlation lengths ξ of the velocity fluctuations. The dashed lines are guides to the eye.

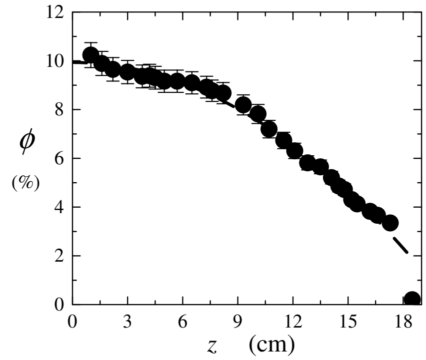

FIG. 3. Particle volume fraction ϕ as a function of height z. The dashed line is a guide to the eye.

The velocity fluctuations, the volume fractions, and the correlation lengths all depend on height. How do we understand these surprising results and why does steady state sedimentation display these properties? The meaning of steady state is that at all points in the particle column the average volume fraction is constant in time, i.e., $\partial\phi(z)/\partial t = 0$. This condition can be recast in terms of particle fluxes using the conservation of mass, or continuity, equation $\partial\phi(z)/\partial t = -\nabla \cdot \mathbf{j(z)}$ [10], where $\mathbf{j(z)} = \phi(z)\mathbf{v(z)}$, and $\mathbf{v(z)}$ is a locally coarse grained velocity. Expanding the particle flux to first order in fluctuations, $\mathbf{j(z)} = \mathbf{j_0(z)} + \boldsymbol{\delta}\mathbf{j(z)}$, and assuming $\partial j_x/\partial x = \partial j_y/\partial y = 0$, and $j(z) = j_z(z)$, the continuity equation becomes

$$\partial\phi(z)/\partial t = -\partial j_0(z)/\partial z - \partial[\delta j(z)]/\partial z. \tag{1}$$

Stability requires that gradients in the local particle fluxes sum to zero, $\partial j_0(z)/\partial z + \partial[\delta j(z)]/\partial z = 0$.

The particle flux gradient due to fluctuations is found by expanding the velocity and concentration fields [and simplifying the notation by writing ϕ, v, ξ, σ_v, and v_{sed} for the height dependent $\phi(z)$, $v(z)$, $\xi(z)$, $\sigma_v(z)$, and $v_{\text{sed}}(z)$] as $\phi \to \phi_0 + \delta\phi$ and $v \to v_0 + \delta v$, yielding $\partial\delta j(z)/\partial z = \partial\langle\phi_0\delta v + \delta\phi v_0 + \delta\phi\delta v\rangle/\partial z$, where $\langle\ldots\rangle$ represents an ensemble average at each height. The first two terms are zero because $\langle\delta\phi\rangle = 0$ and $\langle\delta v\rangle = 0$. The product $\langle\delta\phi\delta v\rangle$ is nonzero because volume fraction and velocity fluctuations are correlated [5]; buoyant forces drive more concentrated regions downwards, $(+\delta\phi)(-\delta v)$, and less concentrated regions upwards, $(-\delta\phi)(+\delta v)$, so that $\delta j(z) < 0$ for all fluctuations. To estimate $\delta\phi$, which we have not directly measured, we use a simple model based upon random statistics [1]. Fluctuations occur in regions of linear size ξ, and contain on average $N_\xi = \phi\xi^3/a^3$ particles, assuming that the particles are randomly distributed. The rms fluctuations are determined as $\sigma_{N_\xi} = \sqrt{N_\xi S(\phi)}$, where $S(\phi)$ accounts for excluded volume effects and is calculated from the Carnahan-Starling equation for hard spheres [10]. The fluctuations in volume fraction are then $\sigma_\phi = \sigma_{N_\xi}(a/\xi)^3 = \sqrt{\phi S(\phi)a^3/\xi^3}$. Approximating the derivative term by $\partial/\partial z \to C_\xi/\xi$, where C_ξ is an adjustable constant expected to be of order unity, we find that the gradient in the flux due to fluctuations is $\partial[\delta j(z)]/\partial z \simeq \langle\delta\phi\delta v\rangle/\xi = -C_\xi\sigma_\phi\sigma_v/\xi$ and

$$\partial[\delta j(z)]/\partial z = -C_\xi\sigma_v\sqrt{S(\phi)\phi a^3/\xi^5}. \tag{2}$$

The negative values of $\delta j(z)$ indicate the coupling of velocity and concentration fluctuations results in a net flux of particles downwards. If left unbalanced, this would cause a stratification of concentration with height and a growing accumulation of particles on the bottom [8]. Clearly, there can be no steady state under this condition.

The zeroth order particle flux, $j_0(z) = \langle\phi_0 v_0\rangle$, excludes the effects of fluctuations and is found from the difference between the mean upward flux due to the pump flow, and the mean downward flux due to sedimentation. In the absence of particle settling, the particles would simply be carried upwards by the pumped fluid velocity, and $j_{\text{pump}} = +\phi_0 v_{\text{pump}}$. With the pump turned off, transport occurs only through sedimentation, and $j_{\text{sed}} = -\phi_0 v_{\text{sed}}$, where v_{sed} is the mean settling velocity. In the presence of both terms, the net flux is given by the difference $j_0 = \phi_0(v_{\text{pump}} - v_{\text{sed}})$. The pump forces liquid upwards at a constant velocity $v_{\text{pump}} = 0.525$ mm/s. To determine v_{sed}, we first stabilize the particle column and then turn off the flow pump to allow the particles to settle. Figure 4 shows that the sedimentation velocities are strongly height dependent. At the top of the column, v_{sed} and v_{pump} are nearly equal, but remarkably, at all other positions $v_{\text{pump}} > v_{\text{sed}}(z)$. Our results for v_{sed} compare well with the Richardson-Zaki prediction [10] using the concentration profile $\phi(z)$ in Fig. 3, although some deviations occur near the bottom. Defining the velocity difference as the excess velocity $v_{\text{ex}} \equiv v_{\text{pump}} - v_{\text{sed}}$, and using $\phi = \langle\phi_0 + \delta\phi\rangle = \phi_0$ we find

$$\partial j_o(z)/\partial z = \partial(\phi v_{\text{ex}})/\partial z. \tag{3}$$

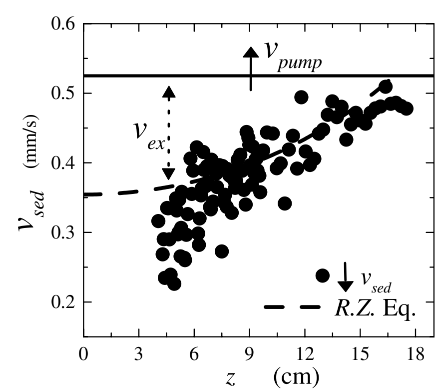

FIG. 4. Local sedimentation velocities v_{sed}, measured after the fluidizing pump is turned off, as a function of height z. The dashed line is the Richardson-Zaki (RZ) prediction $v_{\text{sed}}(\phi) = v_{\text{St}}(1-\phi)^{5.5}$ using $\phi(z)$ from Fig. 3 and the measured value $v_{\text{St}} = 0.63$ mm/s. The dotted line indicates $v_{\text{ex}} = v_{\text{pump}} - v_{\text{sed}}$, with $v_{\text{pump}} = 0.525$ mm/s.

The positive values of $j_0(z)$ indicate a net particle flux upwards, resulting from pumping fluid upwards through the column at a faster rate than the particles settle.

We have found two sources of particle flux in a stable fluidized bed; one driving particles upwards, one downwards. Can we account for the observed stability of the particle column from these fluxes? To answer this, we explicitly test the stability condition $\partial\phi(z)/\partial t = -\partial j_o(z)/\partial z - \partial[\delta j(z)]/\partial z = 0$ in Eq. (1). We evaluate the flux gradient terms in Eqs. (2) and (3) using polynomial fits of our data for ϕ, ξ and σ_v. The RZ equation [10] is also used to calculate $v_{\mathrm{sed}}(z)$ from our data for $\phi(z)$. The results are shown in Fig. 5. Strikingly, when the two flux gradient terms are summed together, we find that the requirement for steady state, $\partial\phi(z)/\partial t = 0$, is well satisfied at all heights in the particle column. Moreover, Fig. 5 also shows that the net particle flux, obtained from summing the integrals of the two flux gradient terms, i.e., $j_0(z) = \phi v_{\mathrm{ex}}$ and $\delta j(z) = -\int_z^{18.5\,\mathrm{cm}}(\partial[\delta j(z')]/\partial z')dz'$, is also near zero over the entire height of the column. These results show that the particle column is stable because the particle flux downward due to fluctuations is nearly equal and opposite to the flux upwards due to the stratification in ϕv_{ex}. The equality of the flux gradients leads to the stability relation,

$$\partial(\phi v_{\mathrm{ex}})/\partial z = -1.1\sigma_v\sqrt{S(\phi)\phi a^3/\xi^5}, \qquad (4)$$

which shows that the magnitudes of the fluctuations are related to the degree of stratification.

The results presented here suggest a picture for sedimentation that is qualitatively different than that reported in the literature. In many experiments [2–4], simulations [6], and theoretical models [7], fluctuation magnitudes are expressed in terms of volume fractions and settling rates that are assumed to be height independent. This assumption is clearly incompatible with our results in steady state sedimentation shown in Figs. 2–4. Moreover, the stability Eq. (4) shows that well-mixed samples without a stratification, i.e., $\partial(\phi v_{\mathrm{ex}})/\partial z = 0$, are unstable to fluctuations, as Tee *et al.* [8] found. The instability results in a continuously growing concentration gradient, which, as Eq. (2) shows, is due to the fluctuations. Concentration gradients, however, generate a particle flux opposing that from fluctuations. Steady state will occur if the particle stratification becomes large enough that the two flux gradients become equal and opposite.

To our knowledge only two works [8,11] have indicated that fluctuations may be connected to the presence of a stratification. However, both of these focused solely on a stratification of ϕ, and did not consider the (significant) role of the excess velocity v_{ex}.

A key question still remains. How do the fluctuations and the stratification depend upon cell dimensions in steady state sedimentation? The stability relation Eq. (4) requires a knowledge of the stratification in ϕv_{ex} for a determination of $\sigma_\phi\sigma_v/\xi$, but does not allow for an *a priori* calculation of either quantity. Future experiments are planned to explore the cell size dependence in stable fluidized beds, which will help to resolve these issues.

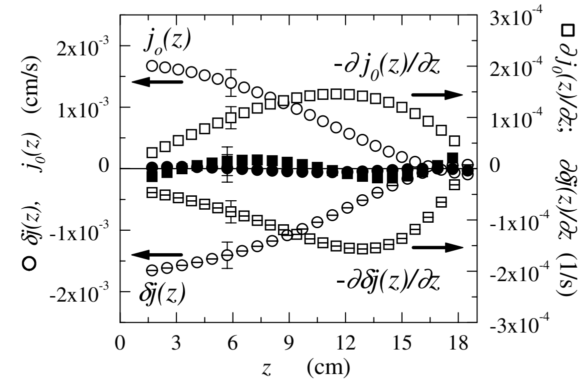

FIG. 5. Stability test: Particle flux gradients $-\partial j_0(z)/\partial z$, $-\partial[\delta j(z)]/\partial z$ (open squares), and their sum $\partial\phi(z)/\partial t$ (closed squares). Particle fluxes $j_0(z)$ and $\delta j(z)$ (open circles), and their sum $j_0(z) + \delta j(z)$ (closed circles). With $C_\xi = 1.1$, the net particle fluxes and their gradients are nearly 0, indicative of a stable, steady state particle column.

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