Experimental study on the dynamics of gas-fluidized beds

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(Received 21 February 2002; revised manuscript received 20 September 2002; published 14 January 2003)

Gas-fluidized fine powders display three regimes of fluidization: solidlike, fluidlike, and bubbling. We investigate, from both macroscopic and local measurements, the transition between these regimes. We show that the transition between the solidlike and the fluidlike regimes takes place along an interval of gas velocities in which transient active regions alternate with transient solid networks. Although in the apparently homogeneous fluidlike regime large amplitude bubbles are not perceived and the bed expands continuously with increasing gas flow, optical probe local measurements show the existence of mesoscale pseudoturbulent structures and short-lived voids, reminiscent of liquid-fluidized beds behavior, and whose characteristic temporal frequency increases with gas velocity. These mesostructures might be responsible for the fast diffusion measured in gas-fluidized beds.

DOI: 10.1103/PhysRevE.67.016303 PACS number(s): 47.55.Kf, 47.55.Mh, 61.43.Gt

An interesting situation displayed by granular materials is that in which fine particles interact strongly with a flowing fluid. A typical fluidized bed consists of a vertical vessel closed at the bottom by a porous plate, a bed of powder particles, and fluid, either gas or liquid, supplied from below. With sufficiently high flow the bed expands in an interval of apparently uniform fluidization. But if the gas flow is further increased over a critical value fast-rising large bubbles of clear fluid are formed and expansion is curtailed [1]. The question on the homogeneity and dynamical structure of fluidized beds and/or suspensions of fine particles and the related problem of particle fluctuations in granular flows is of extraordinary interest in both applied and fundamental research and has recently raised a strong controversy within the physics and engineer community. Experiments show that gas-fluidized beds of fine powders (particle size $d_p \sim 10\ \mu$m) behave as a weak granular solid if the superficial gas velocity v_g does not exceed a certain critical value v_c (larger than the minimum fluidization velocity) [2]. For $v_g > v_c$ the powder displays a fluidlike behavior in a broad interval preceding visible bubbling. The local behavior of gas-fluidized beds remains, however, poorly understood. From measurements of the mixing rate of fluidized powders, an effective diffusion coefficient can be obtained that would imply mean fluctuation particle velocities as large as two orders of magnitude larger than the gas velocity [2]. Thus fast diffusion, also measured by means of nuclear magnetic resonance [3], can only be explained by the presence of mesoscopic coherent motion within the fluidized bed. Examples are known of sedimenting suspensions [4] and liquid fluidized beds [5] where mesoscale pseudoturbulent structures are observed. Numerical simulations based on the discrete element method on fluidized beds of cohesive particles suggest the presence of active structures before large structures typical of bubbling are perceived [7]. The size, time scaling and cell geometry dependence of these well-organized fluctuations is a subject of strong debate nowadays [8]. On the other hand, it has been observed that nonbubbling liquid-fluidized beds manifest short-lived bubblelike voids, suggesting that the distinction between the bubbling and nonbubbling regimes is not absolute [10]. Short-lived mesoscale structures, with the scaling features of turbulence, are also observed in high-velocity gas-particle flows such as riser flows and have been recently predicted by direct integration of the full, nonlinear equations of motion [6]. They have been recognized also in the simulation of quasistatic shear flow of a model granular medium [9]. In this paper we explore the local dynamics of gas-fluidized beds of fine powders with artificially reduced cohesiveness. The control of interparticle attractive forces allows us to explore a wide interval of uniform gas fluidization, as opposed to usual systems of glass beads for which the interval of uniform fluidization is rather short. We look for fluctuations of the local density by means of an optical probe and correlate the results with macroscopic observations.

Commercially available Canon CLC700 toner (yellow) has been tested ($d_p = 7.6 \pm 2.6\ \mu$m and particle density $\rho_p = 1.2$ g/cm^3). Attending to the small particle size we would predict a sticky flow behavior typical of group C powders in Geldart's classification incapable of fluidization. Otherwise, and since interparticle adhesion is reduced by the use of flow controller additives such as silica, this powder gives a region of bubble-free ''homogeneous'' fluidization in which the particles are supported by the drag force of the fluid and the bed expands taking the appearance of a low-viscosity fluid. The addition of silica nanoparticles that become dispersed on toner particle surface decreases interparticle adhesion because silica is considerably harder than polymer and thus the additive increases the contact hardness. Silica also decreases the adhesion force by reducing the typical size of the surface asperities at contact [11]. When the powder is fluidized with dry nitrogen several regimes are inferred from measurements of the yield strength and the effective diffusion coefficient [2]. We look now for further insight in the dynamics of the fluidized bed from measurements of the time evolution of its height (h). In the experiments the powder partially fills a cylindrical vessel (4.42 cm diameter) and rests on a porous filter of sintered metal particles (5 μm pore size). All the measurements are initiated driving the powder into the bubbling regime by means of a sufficiently large gas flow sup-

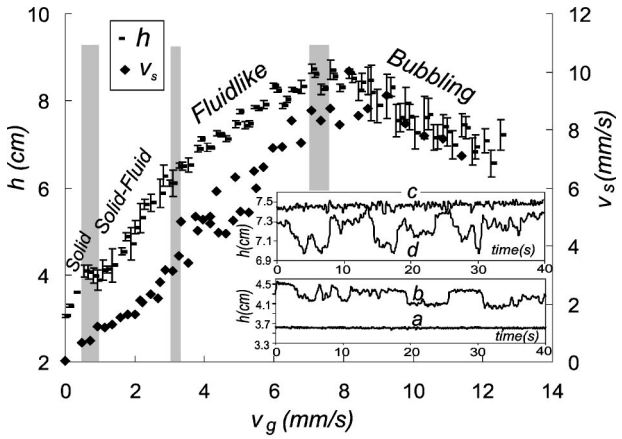

FIG. 1. Fluidized bed height h (left vertical axis) and initial settling velocity v_s (right vertical axis) as a function of the superficial gas velocity v_g for the fluidized bed of toner particles. The insets show time signals of the bed height in the solidlike regime (a, $v_g=0.3$ mm/s), solid-fluid transition (b, $v_g=1.4$ mm/s), fluidlike regime (c, $v_g=4.9$ mm/s), and bubbling regime (d, $v_g=12.5$ mm/s). The error bars in h represent the standard deviation of the time signals of the height.

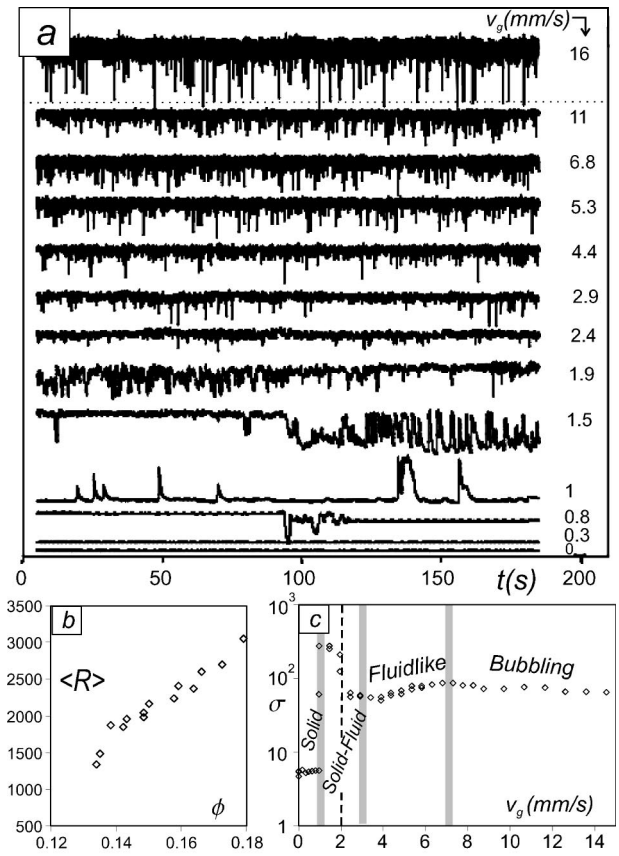

FIG. 2. (a) Time evolution of the reflectance of the fluidized powder (the signal at $v_g=16$ mm/s corresponds to a bubbling bed of glass beads). (b) Average reflectance as a function of the average solid volume fraction. (c) Standard deviation of the time signals as a function of the gas velocity.

plied from below. Once the powder is in the bubbling regime the gas velocity v_g is set down to a value larger that the minimum fluidization velocity ($v_m \simeq 0.1$ mm/s [11]). Then precise measurements of h are taken by an ultrasonic sensor placed on top of the vessel within a time interval of 40 s (time resolution 50 ms). Results are displayed in Fig. 1. For a constant gas velocity $v_g < 0.5$ mm/s the bed is in the solidlike regime, particles are static and thus h remains stable in time. For 0.5 mm/s$< v_g < \sim 1$ mm/s the powder retains a yield strength [2] but superficial transient instabilities appear that cause fluctuations in h. For $v_g > 1$ mm/s, the bed loses the yield strength but the transit to the fluidlike regime is not clear cut. Within the velocity interval 1 mm/s$< v_g < 3$ mm/s metastable solid and fluid regions alternate within the bed, yielding abrupt oscillations of the surface and thus of h. When the entire bed enters the fluidlike regime ($v_g > \sim 3$ mm/s) the free surface returns to a stable state and h increases steadily up with v_g until $v_g \sim 7$ mm/s. At $v_g \sim 7$ mm/s the height reaches its maximum value corresponding to the minimum value of the solid volume fraction that the two phase system can sustain, $\phi = \phi_b \simeq 0.13$. Then a large part of the gas is trapped by bubbles from the dense phase and h decreases if v_g is increased, with strong oscillations of the free surface due to bubbles burst on the free surface. Gas-solids segregation also affects the settling velocity $v_s = dh/dt$ of the powder free surface measured just after shutting down the gas supply (Fig. 1) [12]. In the fluidlike regime $v_s \simeq v_g$, since the mixing of the gas and solid phases is macroscopically homogeneous, whereas in the bubbling regime $v_s < v_g$ due to phase segregation.

To perform the local measurements the sample was held in a 4.7 cm diameter glass cylinder, the base of which is a high-resistance ceramic filter. (Experiments in progress with a rectangular bed indicate that the results do not depend on the particular geometry of the vessel.) The optics system consisted of a fiber optic reflection probe coupled to a spectrometer and a tungsten-halogen light source. The probe is a tight bundle of seven optical fibers (an hexagonal array of six illumination fibers disposed around one read fiber 400 μm in diameter) embedded in a stainless steel ferrule with a 30° window to remove specular reflection effects. (Although unavoidable flow perturbation is introduced by the probe upstream, we expect it to be negligible downstream for its sharp *"V"* shaped tip.) We inserted vertically the probe at 1 cm depth in the center of the fluidized bed and the backscattered light intensity (named hereafter reflectance R) at $\lambda = 550$ nm was acquired each 24 ms. According to our measurements $\langle R \rangle$ in the fluidlike regime decreases with decrease in the average density [Fig. 2(b)]. Furthermore, Mengual *et al.* [13] established, with a similar optical analyzer, that, for an aqueous suspension of latex beads ($d_p \sim 2$ μm) R scaled with the solid volume fraction as $\phi^{0.5}$. Thus, we relate large fluctuations in R to fluctuations in the local density. The characteristic frequency of fluctuations in the pressure due to gas bubbles in most bubbling beds is of the order of 1–5 Hz [14], and therefore sampling with $1/24\ \mathrm{ms}^{-1} \simeq 40$ Hz would be sufficient to detect not only bubbles but also finer structures if present. Figure 2(a) displays typical time series of R. From Figs. 2(a) and 2(c), where the standard deviation σ of R is shown, the transition solidlike-fluidlike

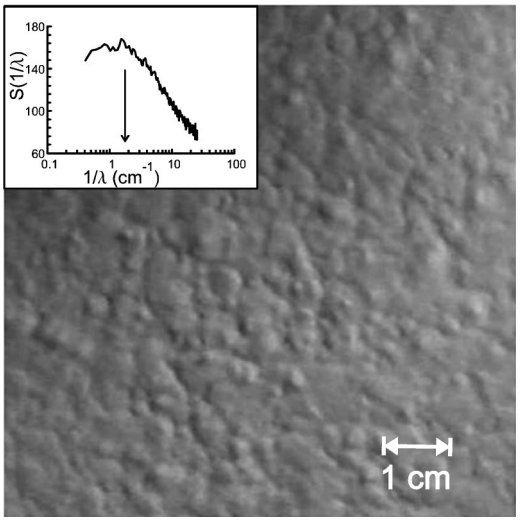

FIG. 3. Upper view of the free surface of the fluidized bed of powder in the fluidlike regime. The inset is an angular average of the two-dimensional fast Fourier transform of the image and shows a characteristic length scale of $\xi \sim 0.6$ cm.

becomes apparent. In the solidlike regime, and due to the presence of stationary solid contact networks, R remains quite constant and the low value of σ is just due to inherent noise of the backscattered light. Since the probe is located close to the free surface we are able to detect in the vicinity of the transition to the fluidlike regime (0.5 mm/s$<v_g<1$ mm/s) the nucleation of strong fluctuations due to transient superficial active regions (see, for example, the time signal at $v_g = 0.8$ mm/s), which are also responsible for fluctuations in the bed height. These surface instabilities cause an abrupt increase of σ. In the interval 1 mm/s$<v_g<2$ mm/s the bed loses its tensile strength, indicating that the transient active regions reach the bulk of the powder. However, we still see short stationary periods due to the developing of transient solid networks. (A similar behavior, reminiscent of a first-order phase transition, was observed by Tsimring *et al.* in a shallow fluidized granular layer [15].) In the transit regime σ reaches a maximum value. Although from the fluctuations in the height measurements we inferred that the fluidlike regime was not fully achieved until the gas velocity surpassed $\simeq 3$ mm/s, the aspect of the time signals and the decrease in σ, suggest that the typical time for the ceasing of surface activity tends to infinity for $v_g > 2$ mm/s. From that point, σ increases continuously with the gas velocity up to a maximum at $v_g \simeq 7$ mm/s corresponding to the onset of bubbling.

In the fluidlike regime mobile particle aggregates can be characterized by the number of aggregated particles $N \simeq 130$ and by the ratio of aggregate size to particle size $k \simeq 6.2$, which we can derive from fitting results on the settling velocity to a modified Richardson-Zaki law [12]. The values of N and k yield an aggregate Reynolds number $\mathrm{Re}_a \sim 0.1$. Thus a fundamental question arises: is this low Reynolds number state a really homogeneous state of uniform local density? The strong fluctuations that we find in R, as compared to the fluctuations for the solidlike homogeneous state, deny that possibility. Otherwise, we must think of a pseudoturbulent state characterized by short-lived mesoscopic structures of the same kind of those observed in other granular flows [4,9] that would originate a rapid diffusion process. Indeed, direct visualization of the fluidized bed free surface (Fig. 3) confirms the existence of well-organized cells on a scale ξ of millimeters. As suggested by Segre *et al.* [4] for liquid suspensions, the mixing mechanism could be driven by aggregates advected to cells, traveling a distance $\sim \xi$ until the correlated cell decays in time, and then becoming entrained in a new cell. This physical picture provides a diffusion process with a mean free path $\sim \xi$ and a diffusion constant $D \sim (\delta v)\xi$, δv being the velocity fluctuations that we assume, following the results of Segre *et al.* [4], comparable to gas velocity. We thus estimate $D \sim 1-10$ mm^2/s, which fits the range of the measured values [2].

We also find in Fig. 2(a) a fraction of points for which the reflectance, and therefore the local density, experiences a big drop. We note that the time signal of the bubbling regime, for which rising gas pockets are visible to the naked eye, shows a similar appearance. In Fig. 2(a) the time signal taken from a bubbling bed of dry glass beads ($d_p \simeq 60$ μm and $\rho_p \simeq 2.5$ g/cm^3) is also depicted. This system of larger particles bubbles for an extra very small increment of the gas velocity above the minimum fluidization velocity [16]. Moreover bubbles, which explode on the surface at an average frequency of $\simeq 5$ Hz, are visibly of larger size and this is reflected by the larger depth of the spikes. Thus we interpret the similar spikes observed in the signals of the fluidlike regime as the footprint of local voids linked to mesoscale structures and captured by the probe. These local voids must develop and disappear continuously, without growing up to a

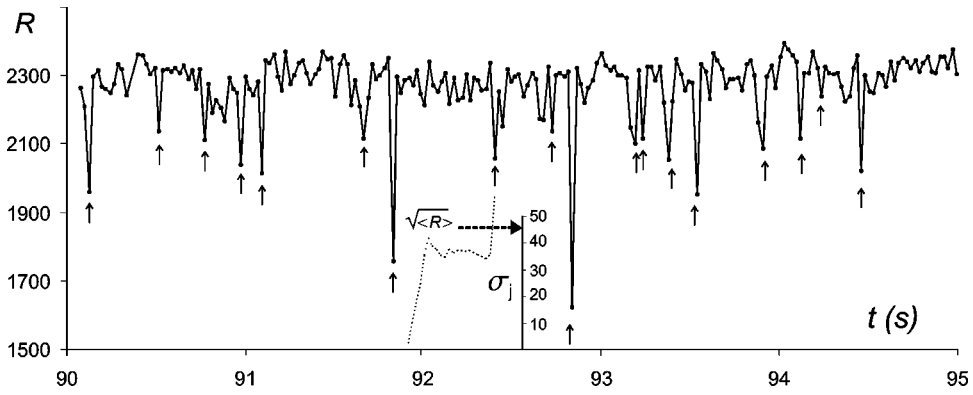

FIG. 4. Zoom of Fig. 2(a) for $v_g = 6.8$ mm/s. The inset shows, for a particular case, the jump of the local standard deviation over the natural deviation when a big drop in the reflectance is detected. The arrows indicate the points identified as local voids by the algorithm.

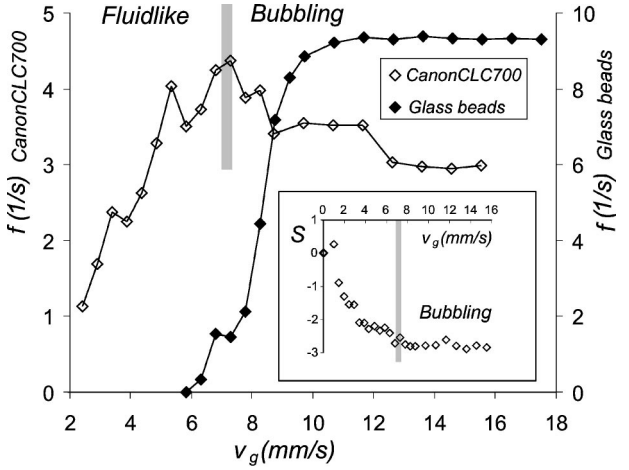

FIG. 5. Average number of local voids detected per unit time f as a function of the superficial gas velocity v_g for the fluidized powder and for the fluidized glass beads. The boundary separating the bubble-free fluidlike and bubbling regimes for the fluidized powder is shown. For the system of glass beads, macroscopic visible bubbles are present at the source of all particle dynamics in the bed. Inset: skewness of the reflectance time signals for the fluidized powder.

macroscopic size. A similar breakup mechanism preventing local fluid pockets from a progressive increase in size is reported in liquid-fluidized beds [10]. In order to have a quantitative information of the fluctuating behavior of the local density in the fluidlike regime, we developed the following algorithm. The expected natural deviation of the light intensity reflected by a homogeneous material is $s=\sqrt{\langle R\rangle}$. Starting from the initial point (which we label $j=0$) we calculate the local average $\langle R\rangle_j(n)=(\Sigma_{i=1}^{n}R_{j+i})/n$ and the local standard deviation $\sigma_j(n)=\sqrt{1/(n-1)\Sigma_{i=1}^{n}[R_{j+i}-\langle R\rangle_j(n)]^2}$, where $n=2,3,\ldots,$ denotes the interval length. As n is increased, $\sigma_j(n)$ increases steadily up until it spikes over the natural deviation for a given $n=n_p$ due to a large drop in the reflectance (Fig. 4). We take then such a point as a representative of a local void and start again the routine from the next point of the time series $(j=n_p+1)$. Results of the average number of voids per unit time (f) are displayed in Fig. 5. It is seen that f increases with gas velocity (as it does σ) up to a maximum value corresponding to the onset of bubbling ($v_g \sim 7$ mm/s). The presence of voids is also reflected on the skewness S of the time signals, which measures their asymmetry. It is seen that S decreases with increasing gas flow, saturating at a minimum ($S_{min}\simeq -3$) when the bubbling gas velocity is reached (Fig. 5, inset). We also show in Fig. 5 the values of f measured for the fluidized bed of glass beads, for which a parallel trend is found. We must note, however, that f shows a monotonic increase for the system of glass beads, whereas the behavior for the fluidized powder is more irregular with a peak at the onset of bubbling. A Fourier analysis of the time signals (Nyquist frequency $\simeq 21$ Hz) does not reveal a characteristic frequency of the fluctuations, in agreement with the conclusions of Letaief *et al.* [17], who found the characteristics of noise, akin to a fractal Brownian motion process, in a bubbling bed in the range of gas velocities close to the minimum fluidization velocity. On the other hand, this behavior contrasts with the strong periodicity usually found on high-velocity macroscale bubbling of deep beds [14].

In conclusion, the macroscopic measurements of the bed height fluctuations have served us to explore the solid and fluidlike regimes of a gas fluidized bed of fine powder. The transition to the fluidlike regime reminds of the characteristics of a first-order transition. Local measurements of backscattered light and direct visualization of the free surface show that, in spite of the uniform smooth expansion exhibited in the fluidlike regime, pseudoturbulent mesoscale structures with short-lived local voids must be present. This behavior is strikingly similar to the recently reported behavior of liquid-fluidized beds with a Froude number $\mathrm{Fr}=v^2/(gd_p)\sim 10^{-2}$, where v is the bubbling fluid velocity and g is the gravity field. In fact, we may estimate for our gas fluidized beds $\mathrm{Fr}\sim 10^{-1}-10^{-2}$ using the effective size of aggregates, in contrast to gas fluidized beds of coarse beads for which $\mathrm{Fr}>1$ at the bubbling point [18]. As the gas flow is increased, the number of local voids detected per unit time increases until large amplitude bubbles are formed and a clear segregation of gas and solid phases occurs. We then recover the usual bubbly regime observed in gas fluidization of larger particles. Even though mesoscale structures do not grow into fully developed bubbles, their mere presence certainly questions the applicability of hydrodynamic linear or weakly nonlinear stability analyses to predict the onset of bubbling.

We are glad to acknowledge R. Jackson for his fruitful comments on our work and S. Sundaresan for sharing his unpublished results with us. This research was supported by Xerox Foundation, Spanish Government Agency Ministerio de Ciencia y Tecnologia (DGES) under Contract No. BMF2000-1056, and Accion Integrada Hispano-Francesa HF2001-0127.

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