

## THE ROLE OF COHESIVE FORCES ON THE FLUIDIZATION BEHAVIOR OF FINE POWDERS

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Gas-fluidized beds display three main regimes of behavior: bubbling, fluidlike, and solidlike. At large gas velocities there is a continuous bubbling process in which the bed loses memory of previous states. If the gas velocity is decreased below a critical value bubbles disappear and the bed acts as a low viscosity liquid. Due to interparticle attractive forces, particles aggregate and aggregates interact hydrodynamically with the gas. By means of settling experiments we characterize aggregates. Optical probe measurements reveal the existence of mesoscale pseudoturbulent structures, responsible for an enhanced effective diffusion process, and short-lived voids. As the gas velocity is decreased there comes a point at which aggregates are brought together to a loose packing structure and the bed gets a solidlike appearance. The extension of the fluidlike regime decreases as particle size is increased and shrinks to zero when the particle weight is comparable to the interparticle attractive force.

In the past few decades there has been a strong debate about the structure of stable gas-fluidized beds of powders. R. Jackson [1] proposed a hydrodynamic criterion for the stability of a fluidized bed. Since then theoretical and numerical works have appeared that put emphasis on the hydrodynamic interaction between the solid and the fluid phases as a keystone for the stable structure of a gas-fluidized bed [2]. One decade after the work of Jackson, K. Rietema [3] suggested that interparticle cohesive forces were the responsible for the stabilization of the fluidized bed. These contact forces were assumed to give an effective elastic modulus to the bed that could stabilize the system against small disturbances. In that state the bed would behave like a weak solid rather than a fluid. Menon and Durian [4] reported on measurements by diffusion-wave spectroscopy of the reflection of laser light from fluidized glass beads. They found no fluctuations in the interval of uniform fluidization. Fluctuations were initiated by the instability to bubbling. This finding led them to the conclusion that "the uniformly fluidized state was a completely static state". Remarkably, in these experiments the powders used consisted of particles larger than  $\sim 50\mu\text{m}$ . Smaller particles are very cohesive and cannot usually be fluidized in the conventional sense; they form channels, or lift as a plug in the bed. For particles  $> 50\mu\text{m}$  however the interval of uniform fluidization is very short or even absent, gas bubbles form at or slightly above the minimum gas fluidizing velocity and bed expansion is small. We have used xerographic toners with typical particle size  $d_p \sim 10\mu\text{m}$ . The reduction of cohesiveness by means of surface additives allows for a wide interval of non-bubbling fluidization and with a considerable bed expansion (with up to  $\sim 0.9$  of free volume), enabling us to explore in detail the transitions between the different regimes of behavior exhibited by our gas-fluidized beds.

The first of our series of experiments [5] demonstrated that both regimes, the solidlike, for gas velocities  $V_g$  below a critical value  $V_c$ , and the fluidlike, for  $V_g > V_c$ , could be found in a non-bubbling gas-fluidized bed. In the solidlike regime, the fluidized color samples could not mix. Toner particles remained in a stationary static state. Accordingly we could measure a non-vanishing tensile strength. A confirmation of the existence of a mechanical strength could be obtained by slowly tilting the fluidized bed. In agreement with Rietema's experiments the bed remained stable while the top surface was tilted as well. When the gas velocity was increased above the critical value  $V_c$ , a radically different behavior was found. In this fluidlike regime there was no yield stress. Stresses in this state were carried by particle collisions due to fluctuations in their motion. The powder then took on many of the properties of a low-viscosity liquid, its upper surface remaining horizontal when the container was tilted. Color samples mixed successfully and the mixing process could be described by an effective self-diffusion coefficient. The fluid-to-solid transition became also clear when we investigated the response of the fluidized bed to vertical vibration [6]. In the fluidlike regime the bed expanded reversibly as vibration amplitude was increased. On the contrary in the solidlike regime the bed behaved like a weak granular solid relaxing irreversibly toward a more compact state by eliminating low-density metastable configurations.

When the gas velocity was further increased above a critical value  $V_b > V_c$  large gas bubbles appeared. Even though the only existence of a fluidlike regime denied that the source of macroscopic bubbling could be the loss of mechanical strength, our results merely showed that, for sufficiently small particles, macroscopic bubbling was inhibited by some complex hydrodynamic mechanism over an extended fluidlike interval. In fact careful measurements by Tsinontides and Jackson [7] on glass beads and cracking catalyst of particle size  $> 50\mu\text{m}$  had clearly demonstrated that there was no interval of bubble free expansion beyond the point where the yield strength became zero. We carried out a new series of experiments indicating that the extension of the fluidlike interval shortened as the particle size was increased [8]. Plotting the width of this interval as a function of the granular Bond number  $Bo_g$  of the particles (defined as the ratio of the interparticle attractive force to particle weight) we predicted that the fluidlike interval should shrink to zero for  $Bo_g < \sim 10$ . A rough estimation yielded that for  $d_p > \sim 40\mu\text{m}$  bubbling should be only restrained by the solidlike behavior of the fluidized bed in agreement with Rietema. Our results should therefore serve to put an end to arguments about hydrodynamics versus contact forces as stabilizing mechanisms.

The relationship between the settling velocity in sedimentation  $V_s$  and the particle volume fraction  $\phi$  for fluidized noncohesive spheres has been well described by the phenomenological Richardson-Zaki (R-Z) law,  $V_s = V_{p0}(1 - \phi)^n$ , where  $V_{p0}$  is the settling velocity of a single particle,  $n \sim 5$  (low Reynolds number limit), and  $\phi$  is the particle volume fraction. When we came to measure [9] the sedimentation velocity of nonbubbling fluidized beds just after the gas supply was suddenly stopped, we found that the top free surface settled initially at a constant velocity. Not surprisingly

we found that for solidlike states ( $\phi > \phi_j$ ) the experimental  $V_s$  was lower than the R-Z prediction since settling was hindered by permanent contacts. On the other hand, for fluidlike states ( $\phi < \phi_j$ )  $V_s$  exceeded the R-Z prediction. Indeed fine particles are expected to aggregate since the interparticle attractive force is much larger than particle weight. Our approach was to consider an ideal system of mono-sized aggregates each one consisting of  $N$  particles and with a radius of gyration equal to the hydrodynamic radius  $k$  (relative to particle size). Settling could then be well described by a modified R-Z law  $V_s = V^* (1 - \phi^*)^n$ , where  $V^* = V_{p0} N/k$  is the settling velocity of an individual aggregate and  $\phi^* = \phi k^3/N$  is the volume fraction filled by the aggregates. The fractal dimension of the aggregates was  $D \sim \ln N / \ln k \sim 2.5$  as predicted by the diffusion-limited-aggregation (DLA) model, where self-similar fractal ramified patterns are grown via irreversible sticking of particles in a random motion. The characteristic screening effect of DLA aggregates justified our assumption that the hydrodynamic and the gyration radius of the aggregates are similar. As expected we found that  $N$  and  $k$  decreased as  $Bo_g$  was decreased. The trend suggested that particles should behave as individuals in the limit  $Bo_g \sim 1$ , which is close to the border limiting the existence of a fluidlike interval of fluidization. Thus aggregation seemed to play a main role in preventing the growth of large bubbles along an extended fluidlike interval. As a matter of fact the fluidlike interval also shortened when the size of the aggregates was decreased either by increasing the surface additive concentration or by vibrating the bed, thus breaking aggregates.

The fluidlike regime is an interval of non-bubbling expansion. Nonetheless and since bubbles are well-developed instabilities, we might wonder whether the fluidized bed is really linearly stable in this interval. From our measurements of the effective self-diffusion constant [5] we could infer mean fluctuation velocities for the particles as large as two orders of magnitude greater than the gas velocity. It cannot be conceived any purely hydrodynamic diffusive mechanism by which such huge velocities could be generated in a uniform suspension. We should therefore conclude that fast diffusion must arise from the growth of an instability that does not develop into large visible bubbles. This was confirmed when we looked at the time signals of backscattered light from a fiber optic reflection probe [10], providing us a measure of the local concentration of solids. The results, along with visual observation of the free surface, indicated the existence of mesoscopic structures as well as the development of local voids. The number of local voids detected per unit time increased with gas flow until large amplitude bubbles developed and a clear segregation of gas and solid phases occurred. In the nonbubbling regime the measurements indicated that the local voids growth saturated at a certain height whereas at incipient bubbling the local voids continued to grow up to reach a macroscopic size. This behavior is strikingly similar to the behavior of liquid-fluidized beds of dense beads with a Froude number  $Fr = V_b^2 / (gd_p) \sim 10^{-2}$  [11]. In fact we estimate for our gas fluidized beds  $Fr \sim 10^{-1} - 10^{-2}$  if we use the effective size of aggregates instead of the particle size. This value contrasts with the Froude number of gas-fluidized beds of coarse beads, typically  $Fr > 1$ .

The fluid-to-solid transition parallels non-equilibrium kinetic transitions exhibited by disparate systems such as supercooled liquids, foams, emulsions and colloidal suspensions [12] (usually referred to as jamming). For noncohesive hard spheres  $\phi_j \approx 0.56 - 0.59$  [12], which is comparable to the random loose packing measured in the limit of zero gravitational force (0.56) but is well below the random close packing theoretical limit (0.64). Our fluidized bed experiments [13] indicated that aggregates jam at volume fractions  $\phi_j^*$  independent of the primary particle size, but dependent on the interparticle attractive force  $F$ .  $\phi_j^*$  approached the limit of noncohesive spheres as  $F$  was artificially decreased by means of surface additives. Moreover in the close vicinity of jamming the stress- (volume fraction) relationship could be well fitted to a critical-like functional form  $\sigma \sim (\phi - \phi_j)^\beta$  as predicted by simulations of dry noncohesive spheres, and where  $\beta \sim 1$  is close to the experimental values measured for foams and emulsions. Indeed the ratio of the attractive force to aggregate weight is independent of primary particle size and only slightly above one, which roughly signals the border between cohesive systems and free flowing granular materials. Therefore aggregates might behave effectively as low cohesive spheres also in the weak solid, at least in the close vicinity of jamming. As soon as the consolidation stress became  $\sigma > \sim 20$  Pa (depending on the interparticle force), the experimental data deviated from the power law behavior and crossed over to a logarithmic trend  $\Delta\phi \sim \ln \sigma$ , as typically measured on soils. Like the gelation transition of colloids, the colloidal glass transition and the melting transition of foams, the jamming of our fine powders is extremely sensitive to the applied stress. Our measurements of the local reflectance showed the local nucleation of transient (avalanchelike) fluid structures driving the relaxation time of the system to diverge.

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