



Nanofluidization as affected by vibration and electrostatic fields

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ABSTRACT

In this paper we investigate the behavior of a fluidized bed of silica nanoparticles under the influence of externally applied vibrations and an electrostatic field. We have observed that the application of these fields separately has opposite effects on bed expansion. On one hand, vertical vibrations enhance bed expansion as the vibration intensity is increased up to a critical value. On the other hand, an electrostatic field applied in the horizontal direction, hinders bed expansion. In previous research papers, it has been suggested that the size of nanoparticle agglomerates could be affected either by vibration or by the action of the electric field. Using the modified Richardson–Zaki method to analyze our experimental data we find that vertical vibration tends to decrease the average agglomerate size in agreement with previous research. However, in this work we look further into the physical mechanisms which affect the response of the fluidized bed. Our results suggest that both vibration and the electric field produce a significant perturbation to the flow of agglomerates within the fluidized bed. Vibration transmits a vertical motion to the agglomerates that enhances bed expansion until the vibration velocity becomes of the order of the expected rising velocity of macroscopic bubbles. At this critical point, bubble growth is stimulated by vibration. A horizontal electrostatic field produces a drift of the charged agglomerates toward the walls that gives rise to fluidization heterogeneity and bed collapse. When both fields are simultaneous applied, these opposed effects can be practically compensated.

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1. Introduction

Gas-fluidization of nanoparticles is a subject of great interest to the chemical, pharmaceutical and other industries which use ultra-fine particles in their products due to its potential for dispersing, coating, granulating, mixing and improving gas–solid reaction efficiency. Fluidization is accomplished by passing a gas through a vertically oriented bed of powder. At a certain gas flowrate the drag balances the powder weight per unit area and the powder becomes fluidized. In some cases, as for example in the gas fluidization of silica nanoparticles, smooth fluidization is observed, with extremely high bed expansion and the absence of visible bubbles in a wide interval of gas velocities (Zhu et al., 2005). However, due to the exceedingly large ratio of interparticle attractive force to particle weight, fluidized nanoparticles are observed to form hierarchical fractal structured, highly porous agglomerates of size d^{**} of the order of hundreds of microns (Valverde and Castellanos, 2007). The effect on particle agglomeration of acting forces, such as interparticle attraction, gravity and gas flow shear forces is a matter of active research since there is

a need to understand the powder bulk behavior from fundamental physical interactions.

The expansion of a uniformly fluidized bed of nanoparticles can be well described by the modified Richardson–Zaki (RZ) equation (Valverde et al., 2001a):

$$\frac{v_g}{v_{p0}} = k_a^{D_a-1} (1 - k_a^{3-D_a} \phi)^n \quad (1)$$

where v_g is the superficial gas velocity, ϕ is the particle volume fraction, v_{p0} is the Stokes settling velocity of a single particle, $v_{p0} \simeq (1/18)\rho_p g_0 d_p^2 / \mu$ (viscous regime), g_0 is the gravity acceleration, d_p is the primary particle size, μ is the viscosity of the gas, the exponent n is of the order of 5 in the viscous limit, k_a is the ratio of agglomerate size d^{**} to particle size ($k_a = d^{**}/d_p$), and the agglomerates are assumed to have a fractal dimension $D_a = \ln N_a / \ln k_a$, where N_a is the number of primary nanoparticles in the agglomerate. Eq. (1) has been fitted to experimental data, which has served to infer an average value of the agglomerate size in fluidized beds, which has yielded results in accordance with independent direct experimental observations (Castellanos et al., 2001; Nam et al., 2004; Zhu et al., 2005; Valverde et al., 2008).

Yao et al. (2002) observed that 7–16 nm silica particles formed multi-stage agglomerates (MSA) in the fluidized bed by three steps.

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Firstly, primary nanoparticles were agglomerated into 3D net-like structures (sub-agglomerates). Secondly, the sub-agglomerates formed simple-agglomerates of size d^* in the range 1–100 μm that existed previous to fluidization. When the bed was fluidized, these preexisting simple-agglomerates further joined into complex-agglomerates of size d^{**} of the order of hundreds of microns. The in situ images of fluidized nanoparticle agglomerates obtained by Hakim et al. (2005) offer evidence of a dynamic aggregation behavior. Agglomeration of simple-agglomerates can be thought as the result of a dynamic equilibrium of attraction between the simple-agglomerates and flow shear, that supports the weight of the agglomerates in the gravity field.

If the simple-agglomerates, existing before fluidization, are considered as effective particles, the relative importance between attractive and weight forces can be evaluated by means of the agglomerate Bond number Bo_g^* , defined as the ratio of the attractive force between simple-agglomerates F^* to the weight of the simple-agglomerate $W^* = N_s W_p$, where N_s is the number of primary nanoparticles in the simple-agglomerate, and $W_p = (1/6)\rho_p \pi g_0 d_p^3$ is the weight of a primary nanoparticle. The weight force of the complex-agglomerate, which acts uniformly through the agglomerate body, is compensated by the hydrodynamic friction from the surrounding gas, which acts mainly at its surface due to the effective screening of the gas flow by the agglomerate (De Gennes, 1979; Wiltzius, 1987). As the complex-agglomerate grows in size the local shear force acting on the simple-agglomerates in the outer layer of the complex-agglomerate is increased. Using a spring model (Castellanos et al., 2005), and considering simple-agglomerates as effective particles, this shear force can be estimated as $F_s^* \approx W^*(k^*)^{D_a+2}$, where $k^* = d^{**}/d^*$. Simple-agglomerates will continue adhering to the complex-agglomerate as long as the force of attraction F^* is larger than F_s^* . The balance between these two forces ($F^* = F_s^*$) thus serves to find a limit to the size of complex-agglomerates

$$d^{**} \sim d^*(Bo_g^*)^{1/(D_a+2)} \quad (2)$$

In the absence of humidity, the main attractive force between simple-agglomerates is the van der Waals short ranged force (Krupp, 1967), which can be approximated by (Rietema, 1991)

$$F^* \approx F_{vdW}^* \approx \frac{A_H d_{as}}{24z^2} \quad (3)$$

where A_H is the Hamaker constant, which for most solids is around 10^{-19} J (Visser, 1972), d_{as} is the typical size of surface asperities and z is the minimum intermolecular distance ($z \approx 3\text{--}4 \text{ \AA}$, Rietema, 1991). A value of $d_{as} = 0.2 \mu\text{m}$ is taken by many authors as representative of the asperity size of micron sized particles (Rietema, 1991). In our case the typical asperity size to estimate the van der Waals force between agglomerates should be the typical size of the asperities of the micron-scale agglomerates. In their study on the fluidization behavior of nanoparticle agglomerates of aerogel powders, Chaouki et al. (1985) used also $0.2 \mu\text{m}$ to estimate the inter-agglomerate force. From these values it is calculated $F_{vdW}^* \sim 10 \text{ nN}$. Using a typical value of $d^* = 30 \mu\text{m}$ for the size of the simple-agglomerates, as inferred from the work of Nam et al. (2004), and $D_a = 2.5$, as inferred from settling experiments (Nam et al., 2004; Valverde et al., 2008; Wang et al., 2006a), Eq. (2) predicts $d^{**} \approx 150 \mu\text{m}$ for 10 nm silica nanoparticles ($\rho_p = 2500 \text{ kg/m}^3$). This estimation agrees with the experimental values measured by several researchers for silica nanoparticles using noninvasive laser-based planar imaging techniques (Nam et al., 2004; Zhu et al., 2005; Wang et al., 2006b; Valverde et al., 2008).

Eq. (2) further allows envisaging the role of physical parameters that might change agglomerate size through their effect on either the attractive force or effective acceleration. For example, the presence of humidity enhances the force of attraction, which gives rise to larger agglomerates (Zhu et al., 2005; Hakim et al., 2005). Experimental

observations on centrifugal fluidized beds (CFB) indicate that the mean agglomerate size decreases as the centrifugal acceleration is increased (Matsuda et al., 2004), which can be explained from the increase in the effective acceleration.

The fitting of Eq. (1) to bed expansion data can be a useful tool to investigate the effect of external fields on agglomeration. For example, the expansion of a fluidized bed of micron-sized toner particles, when subjected to vertical vibration, could be attributed, according to Eq. (1), to a decrease in agglomerate size while the fractal dimension remained approximately the same (Valverde et al., 2001b). According to Eq. (2), the size of the complex-agglomerates in a fluidized bed subjected to vertical vibrations of amplitude A and frequency ν would be given by

$$d^{**} \sim \frac{d_0^{**}}{\Lambda^{1/(D_a+2)}} \quad (4)$$

where d_0^{**} is the agglomerate size in the nonvibrated bed, $\Lambda = 1 + A(2\pi\nu)^2/g_0$, and it is assumed that the size of the simple-agglomerates and the fractal dimension are not changed. Using Eq. (4) in the modified RZ equation (Eq. (1)), it is derived that the particle volume fraction of the vibrofluidized bed should be

$$\phi \sim \frac{1 - \Lambda^{(D_a-1)/[n(D_a+2)]}(1 - k_{a0}^{3-D_a} \phi_0)}{k_{a0}^{3-D_a} / \Lambda^{(3-D_a)/(D_a+2)}} \quad (5)$$

where ϕ_0 is the particle volume fraction of the nonvibrated bed and $k_{a0} = d_0^{**}/d_p$. Valverde and Castellanos (2006) showed that Eq. (5) fitted well to the enhanced expansion of vibrofluidized beds of micron-sized toner particles as the effective acceleration was increased up to a critical point at which bubbling instability was stimulated. Albeit, it must be kept in mind that Eq. (1) has a purely empirical origin. Another possible mechanism that may affect the behavior of a fluidized bed, and is not considered by Eq. (5), is the perturbation of particle flow by external fields.

In the present work we will report experimental results on vibrofluidized beds of silica nanoparticles. In the light of the new data the usefulness of Eq. (5) will be investigated. In a second part of our work we will look at the effect on the fluidized bed expansion of an electrostatic field applied in the horizontal direction as recently investigated by Kashyap et al. (2008). A main observation reported in the work of Kashyap et al. (2008) is that the electric field markedly hindered bed expansion. In our work we will report similar observations. It remains, however, to be discussed what effect the electric field has on interparticle forces. Silica nanoparticles would be polarized when the electric field is applied, giving rise to an additional force. However, it is uncertain whether the strength of this force is relevant or how it would affect agglomeration. In our work we will also investigate the effect on the fluidized bed when both fields are applied. These results will highlight the necessity of bringing into discussion the perturbation of agglomerate flow as a possible physical mechanism that affects the response of the fluidized bed to external fields.

2. Experimental setup

In the experiments reported in this work, a cylindrical fluidization cell made of polycarbonate of internal diameter $D = 2.54 \text{ cm}$ and height $H = 16.2 \text{ cm}$ and with a sintered metal gas distributor (pore size $\sim 5 \mu\text{m}$) at its base was utilized. The cell was placed between two square electrodes with 14 cm of side length. The typical distance between the electrodes was 7.6 cm . One of the electrodes was grounded and high voltage (up to 30 kV) was applied to the opposite electrode using a high voltage DC supply (Spellman model HRH 40PN120/OVP/FG) with a range of $0\text{--}40 \text{ kV}$. In this way the behavior

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