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MICROSCOPIC CHARACTERIZATION OF MECHANICALLY ASSISTED FLUIDIZED BEDS

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ABSTRACT

This work focuses on a method to study the effect of two different fluidization assistance methods to overcome the agglomeration of fine particles in fluidized beds. The characterization of the internal structure of the agglomerates is possible with the use of easy-to-measure macroscopic parameters, such as the height of the bed. The validation will be done with in-situ measurements.

INTRODUCTION

Fine particles, which size is smaller than 50 µm, are increasingly applied in a wide range of industrial fields. The large specific surface area of the fine powder can offer a better performance in electronics, medicine or energy storage. For instance, in the pharmaceutical industry 1 µm particles are used in drug powder inhalers for its capability of reaching the alveoli of the lungs and deposit there (1); or in catalysis, where the use of fine catalytic particles leads to higher conversions since more surface is available for the reaction. By the use of fine particles, many processes can be improved. However, further improvement can often be obtained by modifying the surface of the particles by depositing compounds that will enhance the performance of these materials. The gas-phase technique Atomic Layer Deposition (ALD) (2) provides particles with ultrathin coatings of either inorganic, organic or hybrid compounds based on a layer-by-layer growth mechanism (Fig. 1). A set of two reactions takes place in the surface of the particles to form an atomic-level layer; thicker coatings are achieved by repeating the cycle a desired number of times.

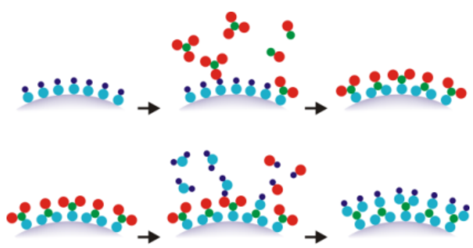

Fig. 1. Mechanism for the deposition of a monolayer using Atomic Layer Deposition.

The production of the core-shell structures can be carried out in a fluidized bed reactor (FBR). Fluidization is an attractive technique to coat particles since large amounts of solids can be processed, and the suspended state that particles experience provides extensive contact between the gas and the solids, which translates into excellent mass and energy transfer in the system. Furthermore, the scalability of fluidized bed reactors enables the production of core-shell particles at industrial scale. Nevertheless, obtaining this ideal behaviour must comply with a homogeneous fluidization of the particles. The large specific area of the fine particles not only improves the efficiency of processes, but increases the tendency to form agglomerates. The fluidization of fine particles, classified by Geldart (3) into the group C powder due to the cohesiveness, will be then hindered by the agglomeration of the particles. That results in the formation of channels, bed cracks and slugging in the column.

The agglomeration can be caused by two interparticle attractive forces: the capillary force and the van der Waals interaction. Fig. 2 shows the value of the forces involved depending on the particle diameter. When decreasing the particle size, the value of the interparticle forces decreases. However, the value of the gravity decreases faster, becoming the attractive interactions the dominant phenomena. The capillary force is stronger, but only if any liquid is present in the system. We use a gas-phase approach for the fluidization and the further coating of the particles, therefore the influence of the capillary in our system is minimum. The agglomeration will be produced by the van der Waals interaction.

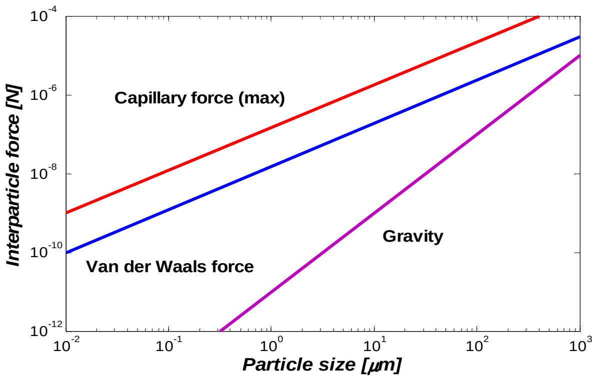

Fig. 2. Comparison of the forces involved in the agglomerate formation depending on particle size. Capillary forces would be significant only if a liquid is present in the system. Figure based on Seville et al. (4).

Homogeneous fluidization of cohesive powders can be achieved by the supply of external energy using fluidization assistance methods. This energy helps in overcoming the interparticle forces, leading to the breakage of the agglomerates. In this work, two different assistance methods will be studied: vibration and agitation (Fig. 3). The effect of the assistance methods is reflected in the bed of particles not only in the macroscopic scale – i.e. lower minimum fluidization velocity or larger bed expansions – but in the microscopic structure. Under an assisted fluidization, the microscopic properties of the agglomerates – e.g. the agglomerate size and the porosity – will change.

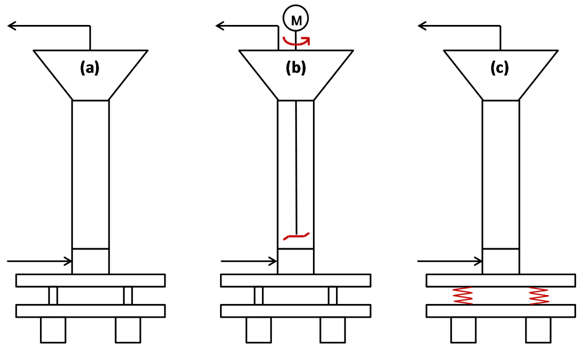

Fig. 3. Scheme of the fluidization assistance methods studied. (a) Non-assisted, (b) stirred and (c) vibrated systems.

Experiments without assistance are used to benchmark the effect of the different assistance methods (Fig 3a). The column is placed in a steady table, and the fluidizing gas is introduced through the bottom. In experiments with stirring, an impeller is introduced in the bed (Fig. 3b), and its rotation would break channels and cracks, producing an enhancement in the fluidization. Alavi et al. (5) found that for rotational speeds larger than 100 rpm, the quality of the fluidization decreased due to the formation of a fixed layer of powder in the walls, obtaining the best results at low rotational speeds. In the vibrated fluidization experiments the column is placed on a shaking table (Fig. 3c). The vertical vibration will produce a pressure wave which propagates along the bed of particles. This pressure wave promotes the breaking of the agglomerates, and at the same time, prevents the gas from forming preferential paths to pass through the bed of particles. Valverde et al. (6) observed an increase in the bed expansion due to the agglomerate breakage in a vibrated fluidized bed experiment.

To determine the effect of the assistance methods, an easy-to-measure variable and a model are combined to correlate the macroscopic properties of the bed of particles with the microscopic structure of the agglomerates. Castellanos et al. (7) extended the Richardson-Zaki (8) empirical model (eq. 1), which was originally developed for the settling of non-cohesive spheres. Castellanos introduced the effect of an agglomerate (eq. 3) instead of a single particle to relate the settling velocity of the agglomerate with its internal structure.

Castellanos measured the time taken by the bed to go from the fluidized to the packed bed state in a collapsing experiment. The settling velocity of the agglomerates was used to estimate the number of particles in an agglomerate N and effective radius R. From these results, the porosity of the agglomerate and the fractal dimension is calculated, obtaining a complete characterization of the agglomerates. Nam (9) proposed a similar approach that relates the expansion of the bed with the microscopic structure of the agglomerates, instead of considering the settling experiment.

$$\frac{v_s}{v_{po}} = (1-\ \phi)^n \qquad \text{(eq. 1)}$$

$$u_{po} = \frac{2}{9} \cdot \frac{(\rho_p - \rho_f) \cdot {r_p}^2 \cdot g}{\mu} \qquad \text{(eq. 2)}$$

$$\frac{v_s}{u_{po}} = \frac{N}{k}(1 - \phi\frac{k^3}{N})^n \qquad \text{(eq. 3)}$$

$$(\frac{u}{u_{po}})^{\frac{1}{n}} = (\frac{N}{k})^{\frac{1}{n}} \cdot (1 - \phi\frac{k^3}{N}) \qquad \text{(eq. 4)}$$

To characterize the effectiveness of the different assistance methods, we measure the height of the bed at different superficial gas velocities, and use the linearized form (eq. 4) of the model proposed by Castellanos to estimate the ratio agglomerate to particle diameter, *k*, and the number of particles per agglomerate, *N*. Besides the comparison of the estimated agglomerate size, parameters such as the bed expansion, minimum fluidization velocity and pressure fluctuations will support the results from the model. This work aims at the development of a method to evaluate the effect of different assistance methods in the fluidization of fine cohesive silica powder.

EXPERIMENTAL

The fluidized bed consists of a 100 mm internal diameter, 500 mm long Perspex column, with a stainless steel SIKA R20 AX distributor plate with a pore size of 23 µm. The column is mounted on a single-motor vibration table Paja PTL 40/40-24. The agitation system is formed by an overhead motor IKA Werk RW 18, and a four-blade stirring paddle. The column was filled with 780 gr of grinded glass with an average particle diameter of 15 µm provided by Kremer Pigmente. Nitrogen at room temperature is used as fluidizing gas. The maximum capacity of the mass flow controller, a Brook 5851E model, is 40 L/min, which is equivalent to a superficial gas velocity of 8,5 cm/s. The pressure drop along the bed was measured with a FSM model DPS differential pressure sensor. Fluctuations in the pressure were also measured with a Kistler 7261 pressure sensor. The minimum fluidization velocity was determined in three different ways. The Ergun equation (10) gives a value of 2,5 mm/s. Analysing the data of the pressure drop versus superficial gas velocity, the value obtained for the minimum fluidization velocity is 4,5 mm/s. However, the visual observation of the experiment does not agree with the calculated and experimental values of the minimum fluidization velocity. At velocities lower than 1 cm/s, the fluidization of the bed is not observed, although the pressure drop equals the weight of the bed of particles. That might be due to an artefact of the vertical vibration that would hold the whole mass of particles without the need of any gas. That would explain that the normalized pressure drop (ratio of the pressure drop to the weight of the bed) equals to 1 at such low superficial gas velocities. At a velocity of 2,5 cm/s homogeneous fluidization of the bed of particles is observed.

RESULTS AND DISCUSSION

To compare the effect of the assistance methods in the fluidization we study first the non-assisted system. Fig. 4a shows the fluidization state at a superficial gas velocity of 8,5 cm/s. The formation of several cracks along the bed of particles is observed, showing that the fluidized state is not achieved.

To study the effect of stirring the bed of particles, we used a 4 cm diameter four-blade impeller placed 5 cm over the distributor plate. This configuration did show a minor improvement in the fluidization state, as can be seen in Fig. 4b. Small channels formed in the area near the wall, and slightly larger bed expansion was achieved when the superficial gas velocity was 8,5 cm/s while the impeller rotated at 75 rpm. However, the bed of particles did not fluidize uniformly. In the center of the column a channel was formed, originating a stream of powder ascending in the column, similar to a fountain. The gas found less resistance to pass through the bed in the region close to the shaft of the impeller. This phenomenon cannot be observed in Fig. 4b.

Finally, we studied the effect of the vibration in the fluidization. Homogeneous fluidization is observed at a superficial gas velocity of 2,5 cm/s and different frequencies of vibration. Visually observation determines a uniform fluidization and a larger bed expansion (Fig. 4c) when comparing with the non-assisted and the stirred fluidization experiments (Fig. 4a,b).

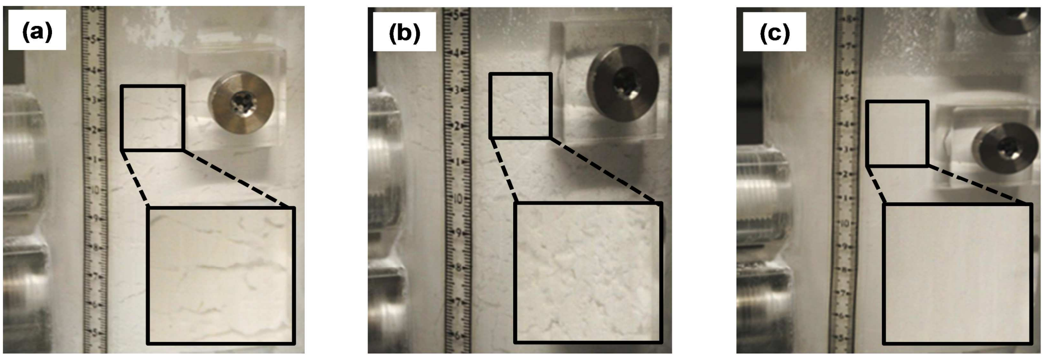

Fig. 4 (a) Picture of the non-assisted fluidization experiment, taken at a superficial gas velocity of 8,5 cm/s. Cracks are visible in the bed. The fluidization state is not achieved with the use of no assistance method. **(b)** Picture of the stirred fluidization experiment, with a four-blade impeller, place at 5 cm over the distributor plate of the bed, rotating at 75 rpm. Small channels are visible near the wall, and a "fountain" of particles is formed in the center, failing to obtain the fluidized state. **(c)** Picture of the vibrated fluidization experiment carried out at a frequency of 25 Hz and a superficial gas velocity of 2,5 cm/s. Homogeneous fluidization is achieved.

We used the vibrated fluidization experiment to study the effect of the different frequencies on the microscopic structure of the agglomerates with the model developed by Castellanos (7). Four vibration frequencies were applied to the system in different experiments: 25, 35, 40 and 45 Hz. A resonance frequency was found at 30 Hz, at which was not possible to perform the measurements. The whole system vibrated violently, giving unstable operation with the risk of damaging the setup. Fig. 5 shows the results of the size (d_{aggl}) and porosity (ε) of the agglomerates at the different vibration frequencies.

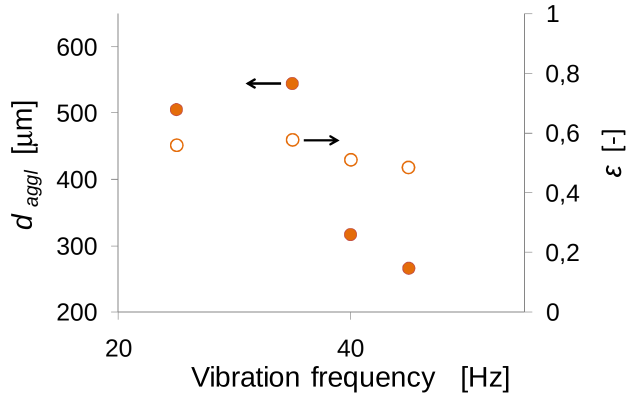

Figure 5. Diameter (circles) and porosity (open circles) of the agglomerates at different vibration frequencies using vibration as fluidization assistance method. The values are calculated using the model of Castellanos (eq. 4).

The results show a decrease in the agglomerate size (Fig. 5 close symbols) when increasing the vibration frequency. At higher frequencies, no remarkable difference in the values of the superficial gas velocity was observed, although it was slightly lower when increasing the frequency. Mawatari et al. (11) defined the parameter “intensity of vibration”, Λ (eq. 5), to describe the force supplied to the system in the form of vibration, which scales directly with the amplitude and frequency of vibration.

$$\Lambda = \frac{A\cdot(2\cdot\pi\cdot f)^2}{g} \qquad \text{(eq. 5)}$$

The amplitude of the vibration in our system increases with the frequency of vibration, ranging from 2 mm at 25 Hz to 7 mm at 45 Hz. Therefore, at larger frequencies of vibration more energy to break the agglomerates is supplied to the system, expecting smaller agglomerate sizes. The porosity of the agglomerates slightly decreases at larger fluidization frequencies. Mawatari et al. (11) found that the porosity of the agglomerates of 20 µm glass beads was barely influenced by the intensity of vibration, reporting a value around 0,6 after solving the force balance of the system. Our results are close to the ones reported by Mawatari, considering that we work with slightly smaller particles, and our particles are not perfectly spherical. These small differences might explain the similarity of the results.

The results from Fig. 5 were obtained with the model of (eq. 4), using the parameter *n* equal to 5,6 as suggested by Castellanos (7). There is controversy in literature about which value should this coefficient take. Nam et al. (9) considered a value equal to 5; Yao et al. (12) used *n* as fitting parameter and found a value of 7,8 after validating the model with TEM pictures. We observed that our results are sensitive to the value given to this empirical parameter, therefore the validation of the model is needed to obtain the value of *n* that best fits our experiments. This will be done in future by recording the agglomerates while fluidizing using the combination of a boroscope, which is inserted into the fluidized bed, and a high speed camera.

CONCLUSIONS

In this work we have studied a method to determine the effect of the assistance methods to improve the fluidization of fine cohesive grinded glass particles. Visual observation indicates the need to use an external supply of energy to the system to obtain a fluid-like behavior of the particles. The bed of particles did not fluidize without assistance. The use of a four-blade stirrer showed a slightly improvement by avoiding the formation of cracks in the bed, but still small channels formed. A different design of the agitator system might produce a larger improvement in the fluidization behavior. Vertical vibration showed to be an effective assistance method to overcome the interparticle forces that hinder the fluidization of the fine particles.

The effect of four different vibration frequencies was evaluated with the model proposed by Castellanos to predict the microscopic properties of the agglomerates. For that, an easy-to-measure parameter, such as the bed expansion at different superficial gas velocities, is used to estimate the size and porosity of the agglomerates. The results of the model determined a decrease in the agglomerate size with increasing the vibration frequency, finding an agreement with the intensity of vibration described by Mawatari. The porosity decreases in a minor range.

The model shows the possibility to characterize microscopically the structure of the agglomerates while fluidizing. This method offers a simple experimentation, is able to estimate the microscopic properties of the agglomerates from easy-to-measure macroscopic variables, is independent of the fluidization assistance method and can be used in scaled systems. However, the validation of the model is crucial. Future work is aimed at the validation of the model to determine a value of the Richardson-Zaki coefficient *n* that best describes our system.

The simplicity of this method opens the possibility to be applied in a wide range of industrial fields; for instance in the production of core-shell particles in fluidized bed reactors with the gas-phase coating technique Atomic Layer Deposition.

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NOTATION

A	Amplitude of the vibration, m
d_{aggl}	Agglomerate size, m
f	Frequency of the vibration, s^{-1}
g	Gravitational constant,$m \cdot s^{-2}$
k	Ratio agglomerate to particle size, -
n	Richardson-Zaki coefficient, -
N	Number of particles in one agglomerate, -
r_p	Radius of the particle, m
u	Superficial gas velocity, $m \cdot s^{-1}$
u_{po}	Stokes settling velocity, $m \cdot s^{-1}$
v_s	Settling velocity of the agglomerates,$m \cdot s^{-1}$
Λ	Intensity of vibration, -
ε	Agglomerate porosity, -
ϕ	Solid fraction of the bed, -
ρ_f	Gas density,$kg \cdot m^{-3}$
ρ_p	Particle density,$kg \cdot m^{-3}$

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