Experimental study on fluidization of micronic powders

Shila Alavi, Brigitte Caussat*

Laboratoire de Génie Chimique/ENSIACET/INPT, UMR CNRS 5503, 5 rue Paulin Talabot, BP1301, 31106 Toulouse Cedex 1, France

Abstract

The fluidization behavior of yttrium oxide (Y_2O_3) powders of high density and micronic diameter belonging to the group C of Geldart's classification has been investigated. Large interparticle forces lead to bed cracking, slugging and channelling, and cause the powder not to fluidize consistently.

Different fluidization technologies have been tested, such as mechanical agitated fluidization, vibrated fluidization and addition of easy-to-fluidize large particles to fine particles. The quality of fluidization has been studied through pressure drop diagrams for decreasing gas velocities and for various fixed bed heights to column diameter ratios.

In the case of stirred fluidization, several stirrer geometries have been tested (helix, turbine, etc.). However, the fluidization has not been satisfactory.

By adding larger particles to fine powders, convenient fluidization conditions have been obtained. An inertia effect proportional to the initial bed weight seems to contribute to fluidization. Some evaluation of interparticle forces governing the tested mixture of fine/large particles has been performed by studying the influence of mass percentage of fine particles on the Hausner ratio and the angle of repose.

Fluidization under vibration allows to partly overcome the adhesion forces between powders. The fluidization behavior has been improved for the highest vibration strengths.

Keywords: Vibrated fluidized bed; Stirring fluidized bed; Fluidization of mixtures; Cohesive powder

1. Introduction

Industrial interest for micronic and even nanometric particles is constantly increasing mainly due to their high specific surface area. One of the essential application domains concerns phosphor micronic particles used as paint pigments or to process flat panel displays (FPD).

Whatever the technology used to synthesize such particles (sol–gel, spray pyrolysis, etc.), a thermal treatment at high temperature (>1100 °C) has very often to be performed to densify and crystallize them, so as their final application properties become optimal. These thermal treatments can hardly be performed in a static mode because of the high sinterability of such particles. Gas–solid fluidization appears then as an interesting solution since this technology naturally leads to isothermal conditions and can reach high productivities, especially if organized in a continuous mode.

It should be noted that the fluidization of fine powders can be limited and even impossible due to the presence of interparticle forces [1]. Geldart proposed that fine powders fall into two categories based on their mean particle size and density [2]. Type A powders fluidize homogenously at the minimum fluidization velocity and expand considerably before the onset of bubbling. Type C powders are extremely cohesive and difficult to fluidize. This difficulty arises because interparticle forces such as strong electrostatic charges and Van der Waals forces are greater than those the fluid can exert on particles. Zhou and Li [3] studied the fluidizability of several fine powders and determined that some particles should be fluidized when gas velocities were in excess of their apparent minimum fluidization velocities.

* Corresponding author. Tel.: +33 534 615211; fax: +33 534 615253.
E-mail address: Brigitte.Caussat@ensiacet.fr (B. Caussat).

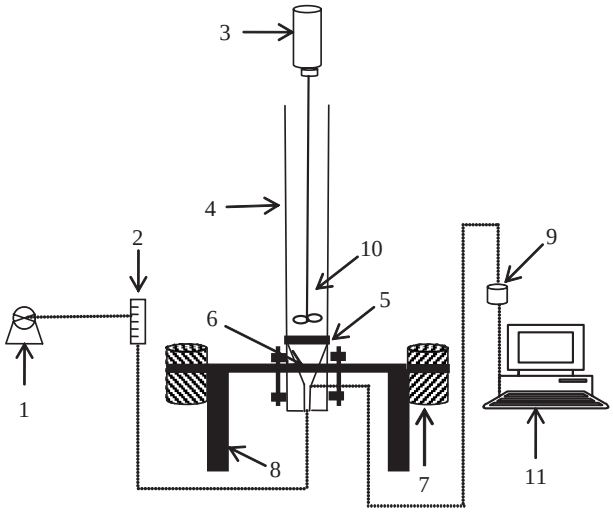
This is due to fact that these Geldart-C particles form agglomerates during fluidization. Other investigators [4] have also analysed the self-agglomeration of such Geldart-C particles. They found that self-agglomeration can be beneficial to the fluidization process in displaying an hydrodynamic behaviour similar to that of Geldart-A particles.

Fluidization of cohesive powders can generally be made possible or at least improved by using mechanical stirrers inside the bed [5]. When particles are not fluidizable by the action of the fluidizing gas alone, high frequency vibrations that break up stable channels can also be helpful [6]. Mori et al. [7] investigated vibro-fluidization of Geldart-C powders and also mixtures of alumina particles of various sizes covering the Geldart-C to Geldart-A groups with Geldart-C group powders, and measured entrainment rates of these mixtures. They found that the entrainment rate depended strongly on mixing ratios. Kage et al. [8] studied the effects of frequency and amplitude of vibrations on the vibro-fluidized bed of fine powders. They found that existence of vibration prevented the progress of agglomeration as vibration amplitude and frequency increased. The addition of coarse particles acting as local turbulence promoters is another way to improve the fluidization of very cohesive particles [9,10]. Dutta and Dullea [11] mixed a small amount of highly dispersed fluidizing aids into Geldart-C powders and found a significant reduction in the cohesivity of powders. They suggested that, by the addition of flow conditioners, interparticle forces or particle interactions could be reduced. In contrast to other methods, the advantage of adding coarse particles to fine powders is that it is unnecessary to get additional equipment or device.

In the present work, the fluidization of yttrium oxide (Y_2O_3) powders belonging to type C of Geldart's classification has been studied by different activated methods of fluidization at ambient temperature. These methods include: mechanical stirring, addition of fluidizable particles to fine powders, vibrated fluidization in cylindrical and conical beds. Our objectives are to develop at high temperature a fluidization process, which is capable to treat group C powders. These Y_2O_3 powders if doped with europium, and crystallized conveniently, may present phosphor properties, which lead to use them as paint pigments or to process flat panel displays (FPD) [12].

2. Experimental features

Fig. 1 shows our central experimental setup, which has been used for testing different activated methods of fluidization. This apparatus consists of a cylindrical Pyrex glass column; its internal diameter and its height are 5 cm and 100 cm, respectively. The transparent column provides a direct visualization of the fluidization quality. The distributor is a porous stainless plate. Dry air has been used as the fluidizing gas; its flow rate has been controlled with a ball flowmeter.



1. Compressor
2. Flow-meter
3. Stirring motor
4. Fluidized bed column
5. Plate distributor
6. Conical bed
7. Vibro-motor
8. Vibrating table
9. Pressure transducer
10. Stirring paddle
11.Computer

Fig. 1. Experimental apparatus.

The total pressure drop across the bed and distributor has been measured by a differential fast response pressure sensor; the high pressure tap has been placed under the distributor. All the measured pressure data have been treated with the DasyLab acquisition system. The pressure drop data have been collected by a data acquisition system with a sampling frequency of 1 Hz. Each pressure drop in the curves presented below corresponds to an average value, which has been calculated from 15 acquisition points. The pressure drop of the bed was obtained by removing the pressure drop of the distributor from the total pressure drop.

Stirred fluidization experiments have been carried out by using a stirred motor and four different types of stirrers as illustrated in Fig. 2. Types a and b represent a pitched blade turbine and a helical paddles stirrer, respectively; both have been installed at the immediate vicinity of the distributor. Types c and d correspond to a comb-form stirrer and a triangle inclined-sharp blades stirrer their heights being equal to 15 and 20 cm, respectively. The overall diameter of the whole stirrers is equal to 0.95 that of the column. The rotational speed has been varied from 30 to 500 rpm.

In conical bed experiments, the porous plate distributor has been replaced by a conical stainless steel piece closed at its base by a porous plate distributor of 1 cm in diameter. The cone angle is 20° and its height is equal to 9.3 cm.

Vibrated fluidization experiments have been performed by fixing the fluidized bed column on a vibrating table as illustrated in Fig. 1. Two vibro-motors are cross-mounted on the opposite sides of the vibrating table. In order to obtain perfect horizontal vibrations, we made a hole on the vibrating table, which allows us to adjust the center gravity of the column at the height of the motors. The vibration

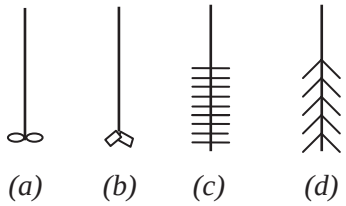


Fig. 2. The various types of stirrers tested (a: helical stirrer, b: pitched turbine blade stirrer, c: comb-form stirrer, d: triangle inclined-sharp blade stirrer).

amplitude has been fixed by varying the eccentric weights on the vibro-motors and its frequency has been controlled by an inverter. The vibrations frequency can be varied from 19 to 25 Hz and their amplitude from 0.5 to 10 mm.

The experiments, which involve adding large particles to fine powders, have been carried out in the fluidization column without exerting any external action. The physical properties of these coarse particles are presented in Table 1. All these particles are hydrophobic. Their terminal velocity is calculated classically by using the Eq. (1) [13].

$$U_t = \frac{d_p^2 g(\rho_p - \rho_g)}{18\mu} \quad (1)$$

Whatever the fluidization method tested, the influence of the H/D parameter has been studied, i.e. the ratio between the fixed bed height and the diameter of the column. This ratio has been varied between 1 and 5.

The minimum fluidization velocity has been classically obtained by the intersection of the fixed bed zone and the fluidization step of the pressure drop curves.

Pressure drop measurements have been performed at decreasing gas velocities for all experiments. These results are presented in terms of normalized pressure drop DP* corresponding to the experimental value divided by the theoretical one (i.e. weight of particles per surface area). The powders used in our experiments are yttrium oxide (Y_2O_3). Its physical properties are given in Table 1 and they are hydrophilic. It should be emphasized that these group C particles cannot be fluidized by a classical fluidization method. The reason is that they form cracks and channels and the pressure drop variation versus gas velocity is quasi-linear.

The weight of elutriated particles has been systematically measured for each series of experiments from the difference between the initial and the final mass of particles in the column.

Table 1
Some physical properties of Y_2O_3 powder and the coarse particles used for mixing with Y_2O_3 powders

Type of particle	d_p (μm)	Bulk packed density (g/cm^3)	Skeleton density (g/cm^3)	U_t (cm/s)	U_{mf} (cm/s)
Yttrium oxide	1.2	5.3	6	0.02	–
Glass	80	1.5	2.7	29	1.8
Sand	300	1.5	2.5	408	8.1
Alumina	290	1.9	3.9	484	8.6
Zirconia	300	3.4	6	926	20.6
Zirconia	500	3.3	6	2497	25.4

2.1. Stirring fluidized bed

Some successful experimental investigations have been reported in the literature, which shows that group C powders, having a mean diameter higher than 20 μm, are fluidizable in mechanically stirred fluidised beds [5]. Some authors have shown that the rotation of the stirrer can break the channels and cracks in forming fluidizable agglomerates of specific dynamic sizes and apparent densities [14].

The effects of the rotational speed and the type of stirrer on the fluidization hydrodynamics of the yttrium oxide channelling bed have been studied.

Fig. 3 presents the evolution of the normalized pressure drop with gas velocity for rotation speeds varying from 30 to 500 rpm, in the case of the triangle inclined-sharp blades stirrer for $H/D=1$.

These experiments have shown that, in the cases of high speeds of rotation (more than 100 rpm), the powders are compressed and pushed towards the wall of the column. This has produced the formation of a fixed cylinder of powders pressed on the walls.

The best results have been obtained for the minimum rotational speed of 30 rpm for all types of tested stirrers. Fig. 4 presents some typical evolutions of the normalized pressure drop for the various types of stirrers tested at 30 rpm, for $H/D=1$.

These results indicate that, for all the stirrer geometries, the fluidization quality has not been satisfactory. The fact that DP* exceeds 1 is due to the strong interparticle forces existing in these beds. Additional experiments have been performed for $H/D=2$ and 4, at high gas velocities (till up to 40 cm/s), but no improvement of fluidization quality has been observed.

Indeed, the forces exerted by the stirrers have not been intense enough to overcome the strong interparticle forces existing in the bed. In the tested conditions ($U<20$ cm/s), any noticeable elutriation has been observed.

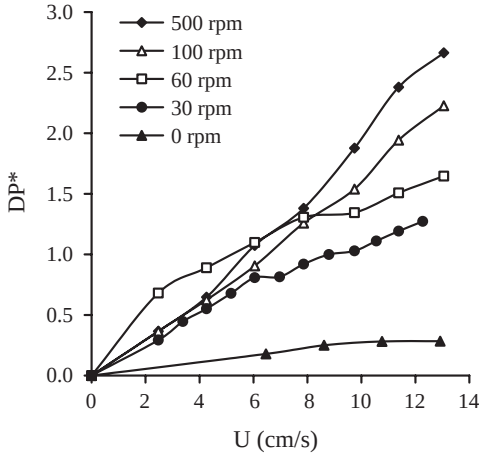


Fig. 3. Variations of the normalized pressure drop with gas velocity for different rotation speeds using the triangle inclined-sharp blades stirrer for $H/D=1$.

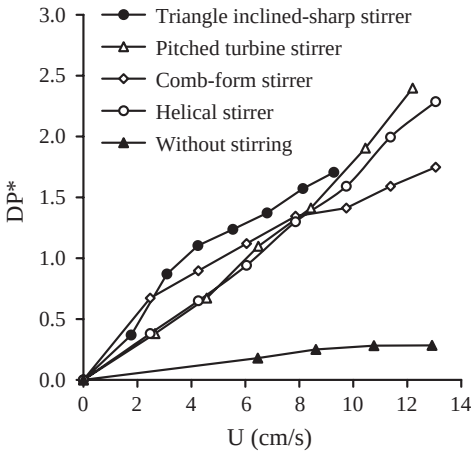


Fig. 4. Variations of the normalized pressure drop with gas velocity using the various stirrers for $H/D=1$.

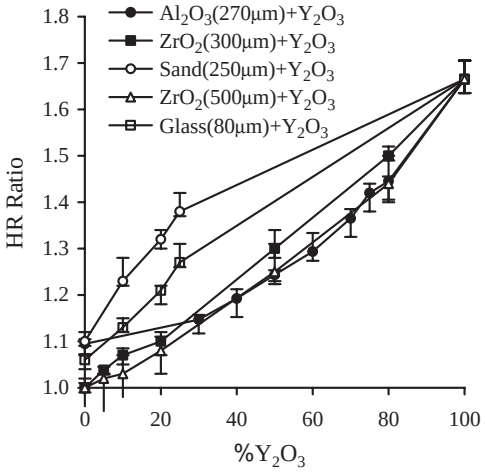


Fig. 5. Effect of the mass percentage of Y_2O_3 in the large/fine particles mixtures on the HR.

2.2. Adding easy-to-fluidize particles

The fluidizability of Geldart-C powders can be improved by adding easy-to-fluidize Geldart-A or -B particles in a conventional fluidized bed. In order to study the effects of density and diameter of the large particles added into the bed of fine powders, different Geldart-A and -B particles have been tested; their physical properties are presented in Table 1.

Two efficient parameters allow following the evolution of the cohesiveness of such mixtures as a function of the mass percentage of fine particles, the Hausner ratio (HR) and the angle of repose. The HR corresponds to the ratio between the tapped bulk density and the loose bulk density. Its range is generally as follows [7]:

- HR > 1.4 : powder is classified in group C,
- HR < 1.2 : powder is classified in group A,
- 1.2 < HR < 1.4: powder may show the behaviour of both groups.

The angle of repose corresponds to the angle between a horizontal plane and the top of a pile of particles. Its value depends on the magnitude of the friction and of the adhesion forces between particles. Powders are then classified as follows.

- $55° < \theta < 70°$: very cohesive powders,
- $45° < \theta < 55°$: cohesive powders,
- $38° < \theta < 45°$: powders with a low flowability,
- $30° < \theta < 38°$: powders with a medium flowability,
- $25° < \theta < 30°$: powders with a high flowability.

The HR and the repose angle have been classically measured using a Hozokawa powder tester apparatus.

The evolutions of the HR and angle of repose with the mass percentage of Y_2O_3 powders in the large/fine particles mixtures are illustrated in Figs. 5 and 6. As previously found by Dutta and Dullea [11], it appears clearly that large particles sensibly reduce interparticle forces in the mixtures. The lowest values have been obtained for zirconia and alumina mixtures.

Fig. 7 shows the variation of the normalized pressure drop with gas velocity for different mass fractions of yttrium oxide in the various mixtures at $H/D=1$. The maximal fluidizable mass percentage of Y_2O_3 for each type of mixtures together and the corresponding minimum fluidization velocity are presented in Table 2. The minimum fluidization velocity for the mixtures is greater than that of pure coarse particles. This is due to the interparticle forces related to the presence of Y_2O_3 fine powders. The best results have been obtained when adding up to 35% in mass of yttrium oxide powders to alumina particles. Beyond this value, the formation of channels and cracks has been unavoidable. Alumina particles have been retained for all the following experiments.

Fig. 8 displays variations of the normalized pressure drop with gas velocity for different mixtures of yttrium and

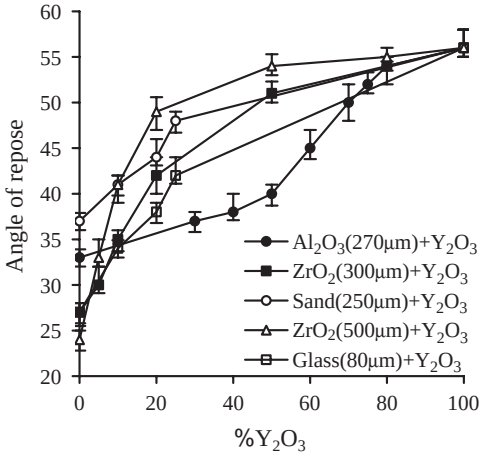


Fig. 6. Effect of the mass percentage of Y_2O_3 in the large/fine particles mixtures on the angle of repose.

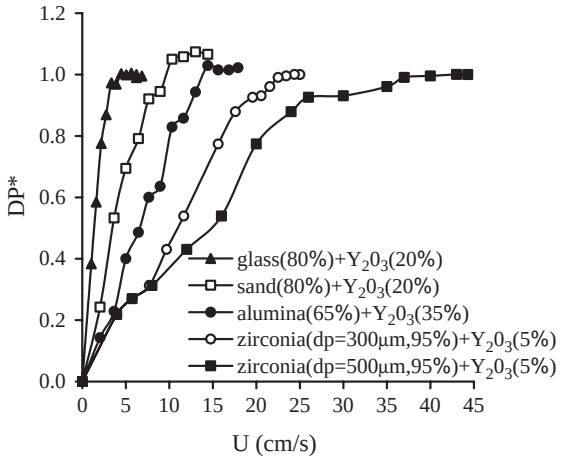


Fig. 7. Variations of the normalized pressure drop with gas velocity for different mixtures with $H/D=1$.

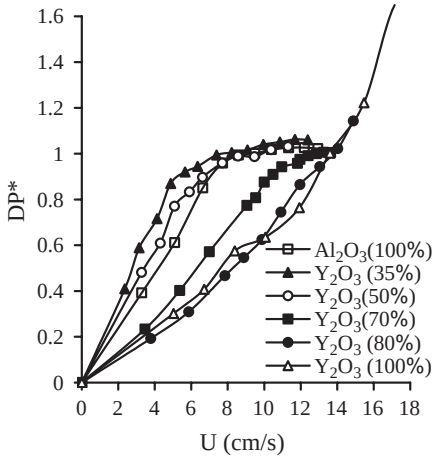


Fig. 8. Variations of the normalized pressure drop with gas velocity for different mixtures of Y_2O_3 and Al_2O_3 with $H/D=4$.

alumina for $H/D=4$; the fluidization of the mixtures has been observed till up to 70% in mass of Y_2O_3 powders.

Consequently, an inertia effect proportional to the initial bed weight seems to contribute to fluidization. It is worth noting that the minimum fluidization velocity for these mixtures appears to be a function of both the mass percentage of fine powders and the H/D ratio.

The influence of the H/D ratio has then been more accurately studied. Fig. 9 shows the evolutions of the normalized pressure drop with gas velocity for mixtures of yttrium oxide and alumina at different H/D values. It is clear that the fluidization quality increases with the increase of H/D ratio up to $H/D=4$. Beyond this value, the fluidization quality decreases, since the normalized pressure drop does not reach the step equal to 1. The intrinsic reasons of such a behavior remain unknown.

Table 2 shows the mass percentage of elutriated particles for the various mixtures at the end of each series of experiments for $H/D=1$. For each mixture, the larger and the denser the coarse particles, the higher their minimum fluidization velocity will be. As the minimum velocity of coarse particles is much larger than the terminal velocities of fine powders, the fine powders are largely entrained in the gas flow when the mixture is fluidized. In the case of adding glass, sand and alumina particles to yttrium oxide powder, the results of elutriation remain acceptable.

Table 2
Experimental results when adding coarse particles for $H/D=1$

Type of particles in mixture with Y_2O_3	Glass	Sand	Alumina	Zirconia (300 μm)	Zirconia (500 μm)
Maximal fluidizable mass percentage of Y_2O_3 in each mixture (%)	20	20	35	5	5
U_{mf} of the mixture (cm/s)	2.9	8.8	13.7	20.8	25.6
Mass percentage of elutriated powders (%)	<1	6	4	12	25

2.3. Vibrated-fluidized bed

Vibrated-fluidization is one of the most efficient methods to improve the fluidization of type C powders. The added vibration energy can overcome the adhesive forces between powders and improve fluidization hydrodynamics.

The effect of high frequency horizontal vibrations on the fluidization of yttrium oxide powders at different vibration strengths has been studied. Vibration strength can be defined as the ratio of acceleration of vibration to that of gravity, as follows:

$$\Gamma = \frac{A(2\pi f)^2}{g}. \quad (2)$$

The frequency of vibration has been varied from 17 Hz to 25 Hz. Several qualitative experiments have been performed at different amplitudes of 2, 3.5, 4.5 and 6 mm. The best results have been obtained at the amplitude of 3.5 mm. This is the reason why the amplitude of vibration has been kept constant at 3.5 mm during the experiments. The direction of

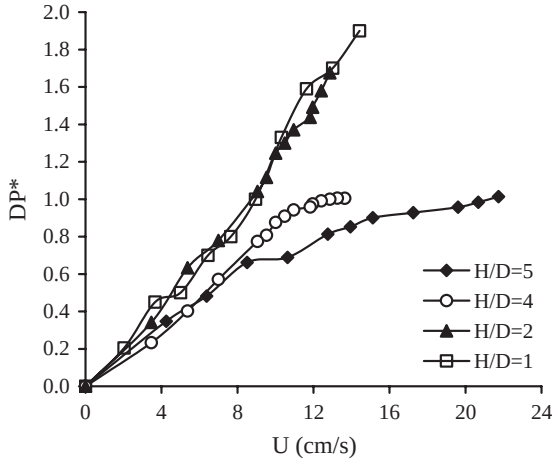


Fig. 9. Variations of the normalized pressure drop with gas velocity for mixtures of Y_2O_3 (30%) and Al_2O_3 (70%) for different H/D ratios.

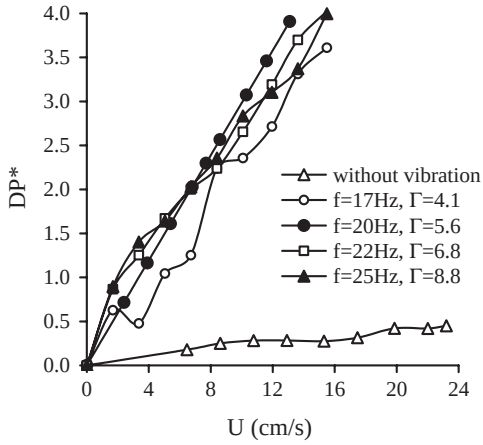


Fig. 10. Variations of the normalized pressure drop with gas velocity for $H/D=1$, in cylindrical-vibrated bed.

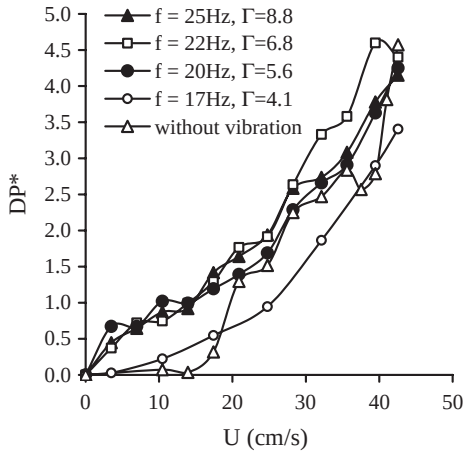


Fig. 12. Variations of the normalized pressure drop with gas velocity for $H=7$ cm in vibrated-conical bed.

vibration has been kept fully horizontal. The effects of the frequency and the H/D ratio in a cylindrical and a conical bed have been analysed.

2.3.1. Cylindrical-vibrated fluidized bed

The variation of the normalized pressure drop with gas velocity for different frequencies at $H/D=1$ are shown in Fig. 10. These curves clearly indicate that convenient fluidization conditions have not been obtained. For the lowest frequencies, thin films of particles adhere to the column walls and, for higher frequencies, agglomerates are easily formed at the upper surface of the bed. The same results obtained at $H/D=4$ are presented in Fig. 11. The fluidization quality has been improved at high frequencies ($f=25$ and 22 Hz) and vibration strengths ($\Gamma=8.8$ and 6.8). The positive effect of an increase of the H/D ratio on the vibrated fluidization of yttrium oxide is evident by comparing Figs. 10 and 11.

It seems that a decrease in the interparticle forces due to inertia effects (as discussed previously) leads to a more breakable bed structure. The energy brought by vibration can partly overcome these lower interparticle forces and improve the fluidization quality.

It is worth to note that the use of vibrations presents the additional advantage of reducing the entrainment of powders even at high gas velocities in comparison with the method of coarse particle addition.

2.3.2. Vibrated-conical bed

At first, some qualitative experiments have been performed to find the optimum fixed bed height of powder to prevent channelling and slugging during fluidization. So two different heights ($H=7$ and 9 cm) have been studied.

The variations of the normalized pressure drop against gas velocity are presented in Figs. 12 and 13.

It should be noted that for $H=7$ cm the results with and without vibrations show that a convenient fluidization has not been achieved. However, in the case of $H=9$ cm, the results of fluidization with vibration, especially at high frequencies ($f=22$ and 25 Hz) are more satisfactory than without vibration. So, another time, an increase of the fixed bed height has contributed to fluidization. In both geo-

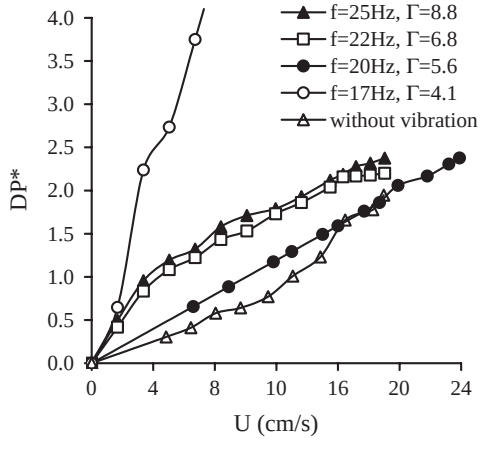


Fig. 11. Variations of the normalized pressure drop with gas velocity for $H/D=4$, in cylindrical-vibrated bed.

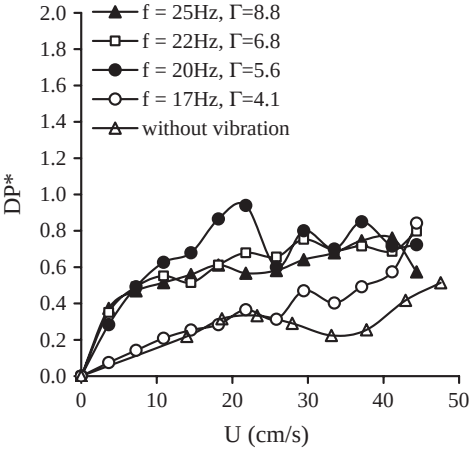


Fig. 13. Variations of the normalized pressure drop with gas velocity for $H=9$ cm in vibrated-conical bed.

metries (cylindrical and conical beds), the best improvements in fluidization quality have been observed at high frequencies and vibration strengths.

3. Conclusions

Fluidization of yttrium oxide (Y_2O_3) powders belonging to group C of Geldart's classification by mechanical stirring has not been satisfactory, particles being compacted on the walls under the rotation of the various stirrers tested.

A good fluidization quality has been obtained by adding coarse alumina particles to the yttrium oxide powders. The measurements of the Hausner ratio and the angle of repose have confirmed the fact that the coarse particles allow fluidization by highly reducing interparticle forces. A maximum amount of 70% in mass of fine particles in the mixture has been obtained for $H/D=4$.

The increase of the H/D ratio until $H/D=4$ has contributed to fluidization in both methods of adding the coarse particles and vibrated-fluidized bed. An inertia effect proportional to the initial bed weight seems to reduce the interparticle forces existing in the bed.

In the vibrated-fluidized bed method, improved fluidization conditions have been obtained for the highest vibration strengths and frequencies in a cylindrical fluidized bed. Fluidization results have not been so satisfactory for vibrated conical bed. For an applied stand point, the fluidization quality seems to be sufficient for developing a high temperature fluidization process of micronic powders.

Several technological solutions are available to develop a high temperature fluidization process for densifying and crystallizing yttrium oxide (Y_2O_3) micronic powders.

Nomenclature

A	Amplitude of vibration (m)
D	Diameter of the column (m)
d_p	Mean diameter of the particles (m)
DP*	Normalised pressure drop, i.e. experimental pressure drop to the theoretical pressure drop ratio (weight of particles per cross-sectional area)
f	Frequency (Hz)
g	Acceleration of gravity (m s^{-2})
H	Height of the fixed bed of particles (m)
HR	Hausner ratio
rpm	Round per minute
U	Superficial velocity of gas (m s^{-1})
U_{mf}	Minimum velocity of fluidization (m s^{-1})
U_t	Terminal velocity of fluidization (m s^{-1})
ρ_g	Gas density (kg m^{-3})
ρ_p	Particle density (kg m^{-3})
θ	Angle of repose (°)
Γ	Vibration strength

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