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ROLE OF SOUND VIBRATION DURING AERATION OF NANO-SIZED POWDERS

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ABSTRACT

The behaviour of two different nano-sized powders, Al_2O_3 (40 nm) and SiO_2 (15 nm), during aeration has been investigated in a laboratory scale fluidized bed. The fluidization quality of both powders is very poor without application of acoustic fields even if some bed expansion has been found. The application of acoustic fields of intensities larger than 135 dB and frequencies close to 120 Hz is able to increase the fluidization quality of both powders. Sound is also able to promote an apparent self-fluidization of a relatively thin portion of the upper part of the bed. The possibility that there is an efficient mixing between aggregates during aeration has been highlighted by experiments using a tracer powder.

INTRODUCTION

In recent years, ultrafine powders, in particular nanoparticles, have received widespread attention with regards to the manufacturing of semiconductors, drugs, cosmetics, foods, plastics, catalysts, paints, sunscreens, biomaterial, due to special physical and chemical properties arising from their extremely small primary particle size and very large surface area per unit mass (1, 2, 3). In this framework, it has become increasingly important to understand how these nanoparticles can be handled and processed: only if control of their size can be made, important potential applications are possible (4). Nevertheless, handling and processing of ultrafine powders is very challenging because they are extremely cohesive (5, 6).

In this framework, fluidization technology has been proposed as a promising technique (1, 4, 5, 7) and has become a significant emerging field for NO_x removal through photocatalytic oxidation, Atomic Layer Deposition (ALD) and synthesis of carbon nanotubes (8). In addition, sound assisted fluidization has been indicated as a suitable technique to have a broad control of the size of nanoparticles aggregates (5, 9, 10, 11, 12). However, in all cases, even if a fluidization regime has been obtained, there were indications that cohesive forces between elementary particles continued to play a key role determining the form and size of loose agglomerates that are the pseudo-particles really characterized by a fluidizing state.

In the present paper the behaviour of two different nano-sized powders, Al_2O_3 (40 nm) and SiO_2 (15 nm), during aeration has been investigated. The materials have been characterized as received by means of granulometric and SEM analyses in order to estimate the size of their natural agglomerates. Aeration tests have been carried out in a laboratory scale fluidized bed at ambient temperature and pressure and using nitrogen as fluidizing gas. Pressure drop and bed expansion curves have been obtained in three different experimental conditions: i) aeration with a fluidizing gas; ii) aeration with both a fluidizing gas and the application of acoustic fields of different intensities and frequencies; iii) aeration only with the application of acoustic fields and without any fluidizing gas. The effect of acoustic fields on the possibility of self-fluidization only under application of suitable acoustic fields, i.e. without any fluidizing gas has been investigated too. The possibility that there is an efficient mixing between aggregates during aeration has been highlighted by experiments using a tracer powder. The size of fluidizing nanoparticles agglomerates have been calculated by working out experimental data and by using Richardson-Zaki relation modified on the basis of the fractal structure of agglomerates (13).

EXPERIMENTAL

The experimental apparatus consists of a fluidization column (40 mm ID and 1000 mm in height), made of quartz, equipped with a sintered bronze porous plate gas distributor, a set of filters for the collection of elutriated fines at the column exit, a pressure transducer installed at 0.02 m above the gas distributor to measure the pressure drop across the bed, a sound wave guide at the top of the freeboard, a sound-generation system and a data acquisition set for sound frequencies (f) and intensities (Sound Pressure Level = SPL), pressure drops (ΔP) and superficial gas velocities (u). The sound-generation system consists of a digital signal generator to obtain an electric sine wave of specified frequency whose signal is amplified by means of a power audio amplifier rated up to 40 W. The signal is then sent to an 8 W woofer loudspeaker placed downstream the sound wave guide. Pure dry nitrogen from tanks is used as fluidizing gas. Tests have been carried out at ambient temperature and pressure.

The behaviour of two different nano-sized powders, SiO_2 and Al_2O_3, has been investigated. The properties of the powders are reported in Table I. The size distributions of the powders as received are reported in Fig. 1. Analysis of data shows that both the powders form relatively large agglomerates, with a Sauter diameter of 250 and 190 nm for Al_2O_3 and SiO_2 respectively. Typical aggregates structures, obtained by using a scanning electron microscopy (SEM), are shown in Fig. 2. Analysis of the pictures shows that both the powders form relatively large aggregates even if those formed by SiO_2 are more irregular.

Aeration tests have been performed in correspondence of an initial bed height (H_0) fixed at about 15 cm, corresponding to a bed of about 50 and 20 g for Al_2O_3 and SiO_2 respectively. Table II summarizes the experimental conditions tested. In particular, the ranges of sound intensity and frequency, having some effects on powders aeration, are reported. These ranges are similar to those used for sound assisted fluidization of cohesive Geldart group C powders (14) and nanoparticles (5, 9). Apart from a direct visualization of the bed, bed expansion and pressure drop curves have been obtained and have been worked out to estimate minimum fluidization velocities (u_{mf}) and average sizes of fluidized agglomerates.

Table I. Properties of the materials tested.

	Al_2O_3	SiO_2
Primary particle average size (nm)	**40**	**15**
Primary particle density (kg/m^3)	**3973**	**2200**
Bulk density (kg/m^3)	**273**	**109**

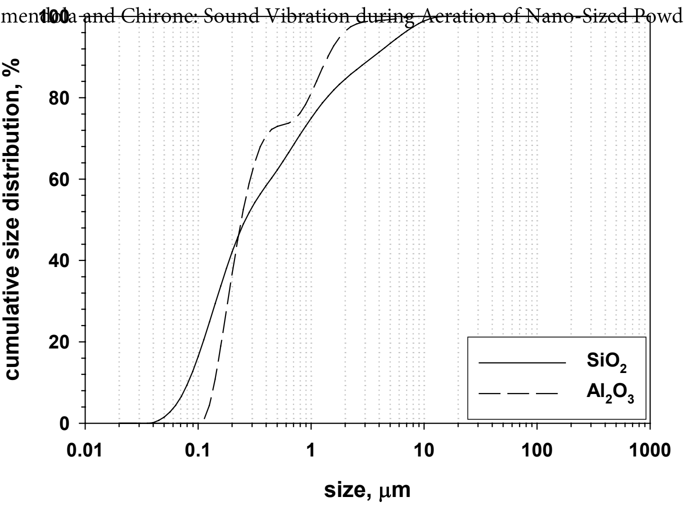
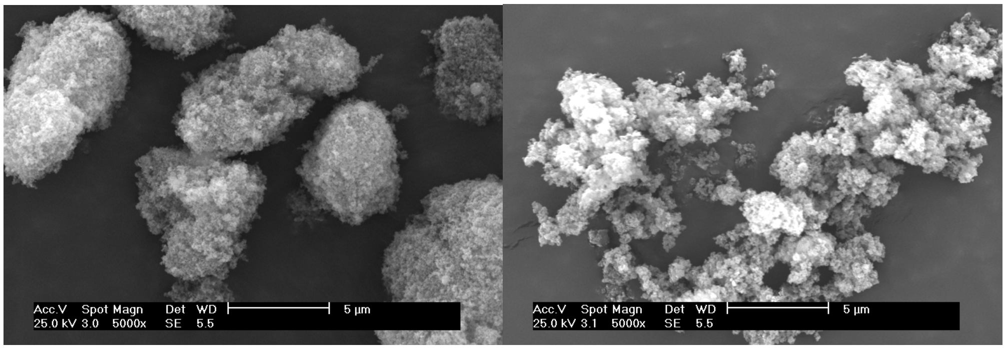


Fig. 1 – Cumulative size distributions of powders.



Al_2O_3 powder

SiO_2 powder

Fig. 2 – SEM pictures of aggregates of Al_2O_3 and SiO_2 powders.

Table II – Experimental conditions.

	Al_2O_3	SiO_2
Operative conditions	**aeration without acoustic fields**	**aeration without acoustic fields**
	aeration with acoustic fields SPL=100-150dB - f=115-125Hz	**aeration with acoustic fields SPL=100-150dB - f=50-300Hz**
	-	**only acoustic fields without aeration SPL=150dB - f=30-120Hz**

RESULTS AND DISCUSSION

Figures 3A and 3B report the dimensionless pressure drop $\Delta P/(W/S)$, where W is the bed weight and S the bed cross section area, and the bed expansion ratio H/H_0, where H is the bed height, as a function of superficial gas velocity obtained for two powders without application of sound. For both the materials, superficial gas velocity has been increased up to a value fixed on the basis of large quantities of powders elutriated from the bed. Analysis of the curves shows that the fluidization quality of both beds is very poor. Even if pressure drop curves approach the unity and some bed expansion has been found, both types of curves are irregular. This is also confirmed by direct observation of the bed.

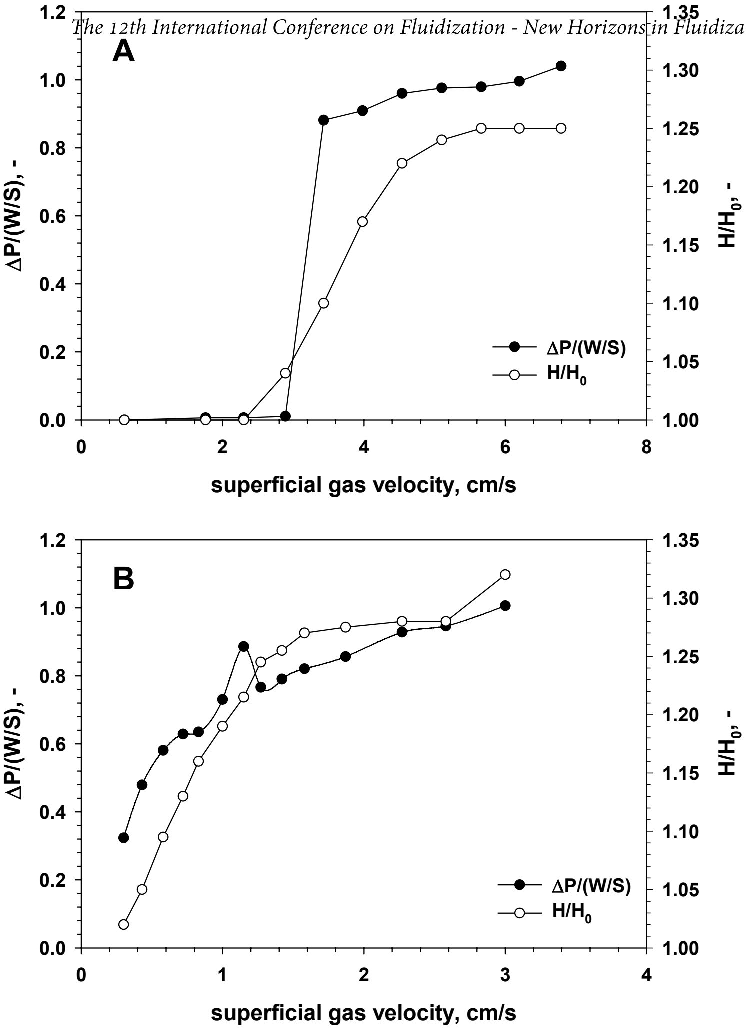


Fig. 3. Dimensionless pressure drop (ΔP/(W/S)) and bed expansion ratio (H/H_0) as a function of superficial gas velocity during aeration without application of sound fields for Al_2O_3 (A) and SiO_2 (B).

Figures 4A and 4B report the dimensionless pressure drop ΔP/(W/S) and the bed expansion ratio H/H_0 as a function of superficial gas velocity obtained for two powders with the application of an acoustic field of SPL=140dB and f=120hz. Also in this case, the superficial gas velocity has been increased up a value fixed on the basis of large quantities of powders elutriated from the bed. Typical ideal fluidization curves have been obtained. Pressure drop data have been collected and analyzed to give the u_{mf} values of 0.01 and 0.39 cm/s for SiO_2 and Al_2O_3 powders respectively. These values are relatively high, about several orders of magnitude higher than the u_{mf} of primary nanoparticles, in spite of the application of an acoustic field. With regards to the bed expansion curves it can be noted that both the powders showed some bed expansion even for superficial velocities lower than the respective u_{mf}. In addition, both the materials showed a homogeneous fluidization like Geldart's Group A powders. The same behaviours have been found by other authors (2) that related such behaviour to the properties of natural aggregates present in the powders and not to the primary particles. This hypothesis is confirmed by the evidence that the found values of u_{mf}, according to Wen and Yu equation (15), would be required to fluidize 80 and 340 μm particles made of materials whose densities is equal to the bulk densities of powders, Al_2O_3 and SiO_2 respectively. Comparison with data obtained without application of an acoustic field indicates that the application of sound waves is required to promote a fluidization state of the powders as a result of break up of the large agglomerates forming the bed into smaller ones. Those agglomerates are in any case significantly larger than both the elementary particles and the so called "natural forming agglomerates". The

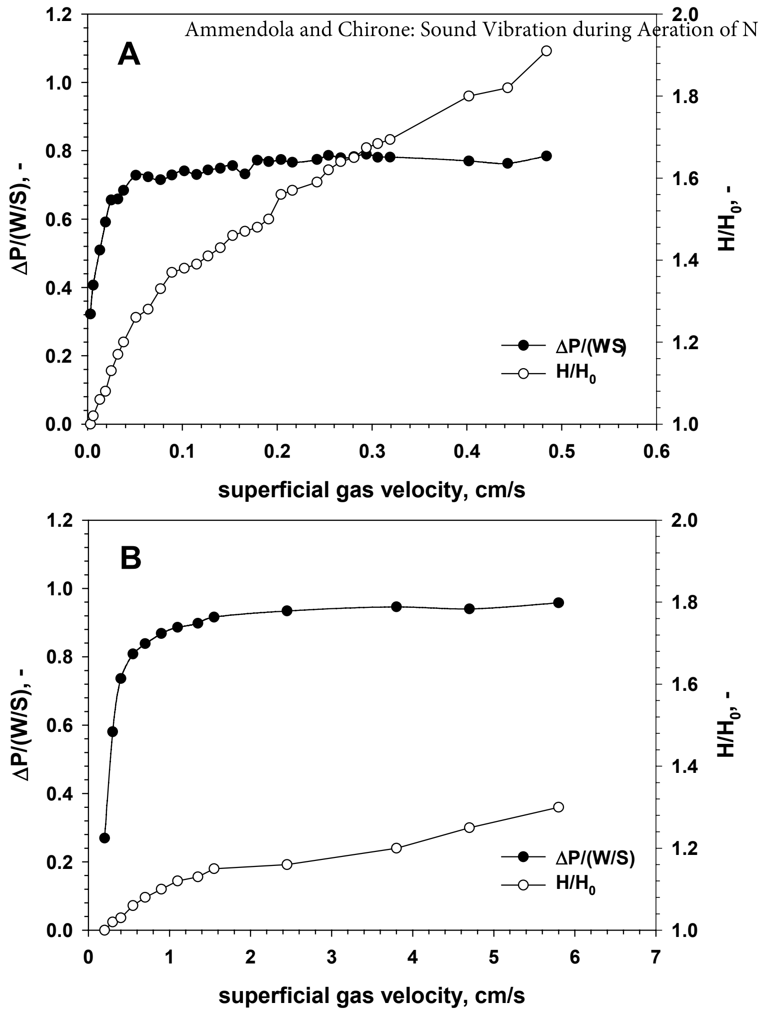


Fig. 4. Dimensionless pressure drop (ΔP/(W/S)) and bed expansion ratio (H/H_0) as a function of superficial gas velocity during aeration with application of a sound field (SPL=140 dB, f=120 Hz) for Al_2O_3 (A) and SiO_2 (B).

application of acoustic fields plays a beneficial role not only in stabilizing the bed aeration but also on elutriation of single fines or smaller aggregates.

Similar pressure drop and bed expansion curves as a function of superficial gas velocity have been obtained for two powders with the application of acoustic fields of different intensities and frequencies. As expected, relatively high sound intensity, larger than 135 dB, is required: intensities smaller than 135 dB did not proved significant influences on bed behaviour.

The effect of sound frequency on u_{mf} is reported in Fig. 5 for both powders at SPL=140 dB. Analysis of the figure shows that the effect of frequency is not monotone and an optimum condition, in terms of a lowest value of u_{mf}, has been found whatever the powder for about f=120hz. The explanation of this trend is not straightforward; it is likely that it is the result of the ability of sound to penetrate the bed as well as to promote agglomerates reduction into a scale which depends also on powder structure.

Because the nanopowders fluidization is characterized by a homogeneous expansion of the bed without bubbles, a combination of Richardson-Zaki equation with fractal analysis of agglomerates, proposed by Nam et al. (13), has been used in order to estimate the agglomerates properties by working out the experimental bed expansion data. In particular, agglomerate sizes of 60 μm and 200 μm and agglomerate densities of 450 kg/m^3 and 300 kg/m^3 have been obtained for Al_2O_3 and SiO_2 powders respectively with reference to the sound assisted fluidization test carried out at SPL=140 dB and f=120 Hz. These sizes are more than 1000 times larger than the primary nanoparticles and also agglomerate densities are 2-3 times larger than the bulk densities of powders. The sizes of fluidized aggregates and their densities obtained have been validated by the evaluation of u_{mf} values by using Wen

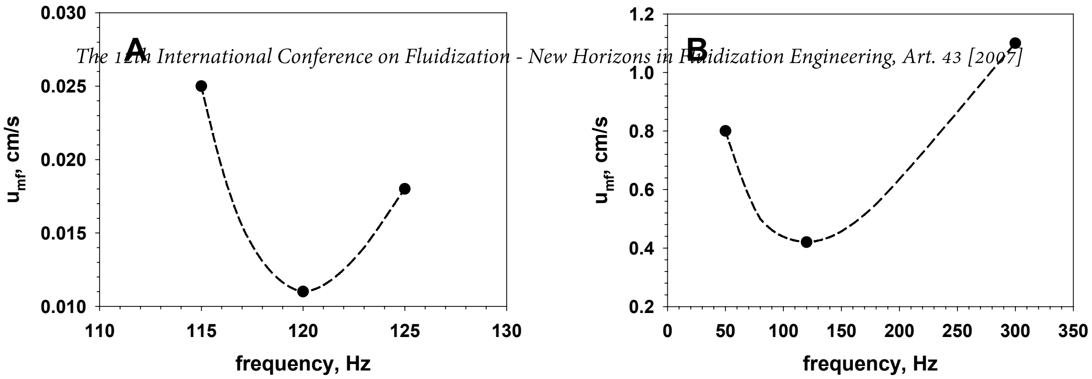
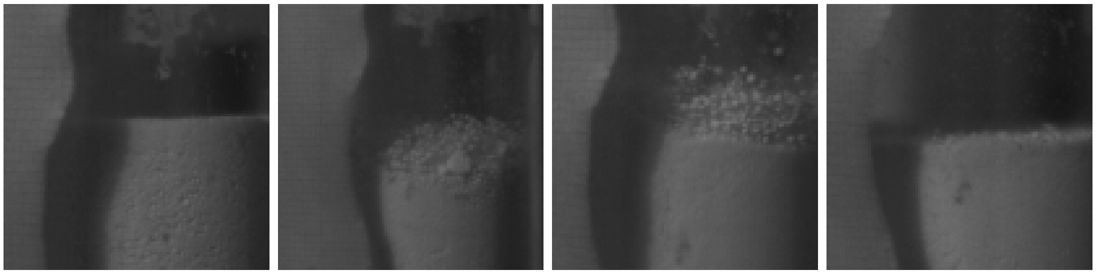


Fig. 5. Minimum fluidization velocity as a function of sound frequency (140 dB) for Al_2O_3 (A) and SiO_2 (B).

and Yu equation (15). The estimated values are close to those obtained from experiments.

The behaviour of a bed of SiO_2 nanoparticles under the effect of application of high intensity (150 dB) acoustic fields of different frequencies and without any fluidizing gas is reported in Fig.6. Analysis of the pictures shows that sound of appropriate frequencies is able to promote an apparent self-fluidization of a relatively thin portion of the upper part of the bed. The range of active frequencies is rather narrow, from 40 to 60 Hz, and smaller than that appropriate to promote a fluidization stage of the powders under sound assisted aeration. In any case relatively large aggregates of particles are formed. Visual observation of the bed also showed that the low as well the high frequencies of sound did not produce self fluidization: for f=30 Hz all the bed moves as a piston while for f=120 Hz an apparent static bed is found.



30 Hz **40 Hz** **60 Hz** **120 Hz**

Fig. 6. Behaviour of a bed of SiO_2 nanoparticles under the effect of application of high intensity (SPL=150 dB) acoustic fields of different frequencies without any fluidizing gas.

The possibility that there is an equilibrium between aggregate breaking and formation during aeration has been highlighted by experiments using a tracer powder. For all the experimental conditions tested, the starting point was obtained by feeding 30 g of the Al_2O_3 nanopowder (white powder) and then 5 g of copper oxide nanopowder (tracer), which forms a black layer on the top of the bed (Fig.7A). The primary particle average size and density of the tracer powder are 33 nm and 6315 kg/m^3 respectively. Fig.7B is relative to 1 minute bed aeration with a nitrogen flow rate corresponding to a superficial gas velocity of 0.46 cm/s. Fig. 7C and Fig. 7D are relative to bed aeration assisted by the application of an acoustic field (SPL=140 dB, f=120 Hz). In all cases the superficial gas velocity used is larger than the u_{mf}

obtained for alumina nanoparticles under sound assisted fluidization. Analysis of the figures shows that without the application of acoustic field no mixing occurs (Fig.7B). Channelling is present and, as a result of stratification of particles of different densities, there is formation of one or more channels with transport of lighter powder on the top of the bed. The application of the acoustic field results into a fluidization regime characterized by a relatively large bed expansion and solid's mixing (Fig.7C). After few minutes the entire bed turned grey and appeared well mixed (Fig.7D). Similar results have been obtained by Nam et al. (13) for a mechanical vibro-fluidization of nanoparticles. Even if both analysis are based only on a qualitative observation of the bed along time sound assisted fluidization seams to be effective as mechanical vibration to promote particle mixing and final large bed expansions.

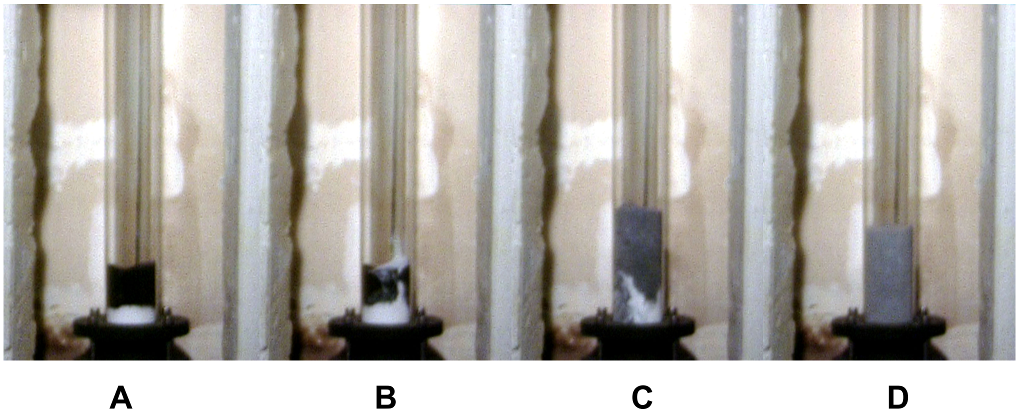


Fig.7. Experiments using a tracer powder.
A - bed made of 30 g of Al_2O_3 (white) and 5 g of CuO (black)
B - bed aerated for 1 min without application of sound (u=0.46 cm/s)
C - bed aerated for 1 min in sound assisted fluidization (u=0.46 cm/s, SPL=140 dB, f=120 Hz)
D - bed aerated for 5 min in sound assisted fluidization (u=0.46 cm/s, SPL=140 dB, f=120 Hz)

CONCLUSIONS

Aeration tests of two different nano-sized powders, Al_2O_3 (40 nm) and SiO_2 (15 nm), have been carried out in a laboratory scale fluidized bed at ambient temperature and pressure and using nitrogen as fluidizing gas. Main results of the study are the following.

- Both the powders form relatively large natural agglomerates with Sauter diameters of 250 and 190 nm for Al_2O_3 and SiO_2 respectively.
- The fluidization quality of both Al_2O_3 and SiO_2 beds is very poor without application of acoustic fields even if some bed expansion has been found. It has been also confirmed by visual observation of the bed and by experiments carried out by using a tracer powder.
- The application of acoustic fields of intensities larger than 135 dB and frequencies close to 120 Hz is able to increase the fluidization quality of both powders as indicated by ideal like pressure drop curves, relatively high bed expansions ($H/H_0=2$) and the occurrence of a homogeneous regime of fluidization. Minimum fluidization velocities of 0.01 and 0.39 cm/s have been obtained at SPL=140 dB and f=120 Hz for SiO_2 and Al_2O_3 powders respectively.

- Agglomerate sizes of 60 μm and 200 μm have been obtained for Al_2O_3 and SiO_2 respectively with reference to the sound assisted fluidization test carried out at SPL=140 dB and f=120 Hz by using a combination of Richardson-Zaki equation and fractal analysis of agglomerates. They are significantly larger than both the elementary particles and the so called "natural forming agglomerates".
- Sound is also able to promote an apparent self-fluidization of a relatively thin portion of the upper part of the SiO_2 bed. The range of active frequencies is rather narrow, from 40 to 60 Hz, and relatively large aggregates of particles are formed.
- The possibility that there is an efficient mixing between aggregates during aeration has been highlighted, even if only on the basis of a qualitative point of view, by experiments using a tracer powder. Again sound assisted fluidization is required to obtain a uniform mixing of the bed.

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