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Sound assisted fluidization of nanoparticle agglomerates

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Abstract

This paper presents some preliminary observations on sound-assisted fluidization of hydrophobic fumed silica nanoparticles (Degussa Aerosil® R974, having a primary particle size of 12 nm) in the form of large 100–400 μm agglomerates. The effect of sound on the fluidization behavior of the nanoparticle agglomerates, including the fluidization regime, the minimum fluidization velocity, the bed pressure drop and the bed expansion has been investigated. It is shown that, with the aid of sound wave excitation at low frequencies, the bed of nanoparticle agglomerates can be readily fluidized and the minimum fluidization velocity is significantly reduced. For example, the minimum fluidization velocity is decreased from 0.14 cm/s in the absence of sound excitation to 0.054 cm/s with the assistance of the sound. In addition, under the influence of sound, channeling or slugging of the bed quickly disappears and the bed expands uniformly. Within a certain range of the sound frequency, typically from 200 to 600 Hz, bubbling fluidization occurs. Both the bed expansion and the bubble characteristics are strongly dependent on the sound frequency and sound pressure level. However, sound has almost no impact on the fluidization, when the sound frequency is extremely high, above 2000 Hz. A relatively high sound pressure level (such as 115 dB) is needed to initiate the fluidization.

Keywords: Fluidization; Nanoparticles; Agglomerate; Sound waves

1. Introduction

Nanoparticle handling and processing technologies have received widespread attention recently because of the increased use of nanoparticles in the manufactures of drugs, cosmetics, foods, plastics, catalysts, and energetic and other advanced materials. In many industrial applications, the nanoparticles are in the form of very large agglomerates due to the strong interactions among the nanoparticles (such as van der Waals forces). A better understanding of the fluidization behavior of nanoparticle agglomerates is therefore of great importance in applications involving mixing, transporting, modifying the surface properties (coating) and downstream processing of nanoparticles to form nanocomposites.

The fluidization of nanoparticle agglomerates has been extensively studied [1–11,22]. It was found that nanoparticle agglomerates can be smoothly fluidized if the gas velocity is increased far above (about several orders of magnitude) the minimum fluidization velocity of primary nanoparticles [1,7]. Furthermore, the minimum superficial gas velocity needed to sustain fluidization of a nanoparticle bed is lower than that required to initiate the fluidization [6,7]. The size of the fluidized nanoparticle agglomerates is typically from about 70 to 700 μm, while the primary particle size ranges from 7 to 500 nm [2,3]. Fluidization results in an extremely high bed expansion, practically no bubbles are observed, and the velocity as a function of voidage around the fluidizing agglomerates obeys the Richardson-Zaki equation [8]. However, a relatively large powder elutriation occurs at the high gas velocities required to fluidize the nanoagglomerates. This loss of particles may hinder the applicability of fluidization of nanoparticle agglomerates in industrial processes.

In addition to conventional gravity-driven fluidization, nanoparticle agglomerates can also be fluidized in rotating or centrifugal fluidized beds [3,4]. In centrifugal fluidization, tiny, indistinct bubbles are observed which become obscure with an increase in rotation speed. The high centrifugal force (much higher than the gravitational force) tends to disrupt the interparticle forces between the nanoparticles forming stable agglomerates, which can be easily fluidized.

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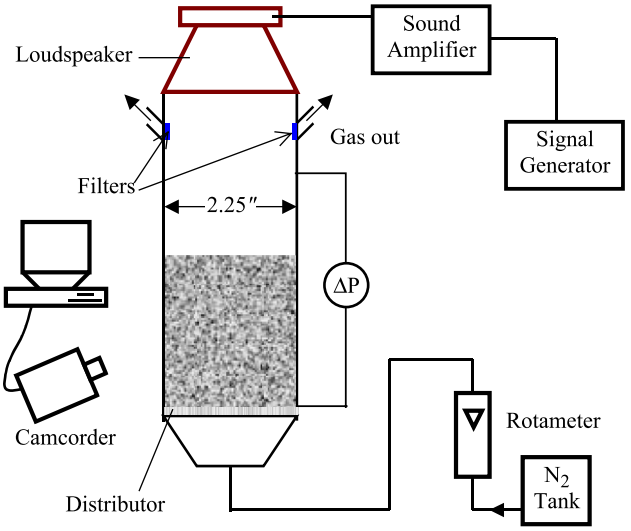


Fig. 1. Schematic diagram of experimental system.

The minimum fluidization velocity of nanoparticle agglomerates in a conventional fluidized bed can be significantly reduced by introducing external excitations to the bed, such as using vertical, sinusoidal vibration [9] or applying an oscillating magnetic field to the nanoparticles that have been premixed with some magnetic particles [10]. With a much-reduced fluidizing gas velocity, hardly any elutriation of the nanoparticles [9,10] was observed. A combination of these excitation methods may further improve the fluidization of nanoparticle agglomerates [11]. In the current study, sound waves are used as an alternative excitation method that is relatively inexpensive, affects the entire particle bed, and does not require any physical contact between the sound generator and the nanoparticles.

Cohesive micron- or even submicron-sized fine particles, typically known as Group C particles, according to the Geldart classification, have been successfully fluidized by the assistance of acoustic fields [12–21]. At a low sound frequency, typically from 50 to 400 Hz, and a high sound pressure level, typically above 110 dB, sound waves can improve the fluidization of fine particles, which otherwise showed intense channeling or slugging rather than fluidization [12]. Standing waves are generated in the experimental column. At a fixed sound pressure level, sound assisted fluidization can only occur within a certain range of low sound frequencies. Channeling is found above and below this frequency range [14]. At the natural frequency of the bed of micron-sized particles, high intensity sound waves lead to reductions in both the minimum bubbling velocity and the minimum fluidization velocities [17]. An increase in sound pressure level may also yield a decrease in bed expansion, an increase in bubble frequency and an increase in bubble size [17,18]. In addition, the high intensity sound can also effectively reduce the elutriation of fine particles [20].

So far, all studies on sound-assisted fluidization have been focused on the fluidization of micron- or submicron-sized particles. No results have been reported on the effects of sound on the fluidization of nanoparticle agglomerates. This paper presents an investigation of sound-assisted fluidization of nanoparticle agglomerates and their fluidization characteristics, which are not only different from those observed using other fluidization methods for nanoparticle agglomerates, but are also different from sound-assisted fluidization of micron- or submicron-sized particles. The effects of sound frequency and sound pressure level on the fluidization behavior, such as the minimum fluidization velocity, bubbling regime, pressure drop across the bed and bed expansion, will all be demonstrated.

2. Experimental method

A schematic diagram of the sound-assisted fluidization system is shown in Fig. 1. The system consists of a fluidized bed of nanoparticle agglomerates, a sound excitation device and a visualization apparatus. The fluidized bed is a vertical transparent column with a distributor at the bottom. The column is a section of acrylic pipe with an inner diameter of 57 mm and a height of 910 mm. The distributor is a sintered metal plate of stainless steel with a thickness of 2 mm and pore size of 20 μm. An ultrafine mesh filter is located at the gas outlet to filter out any elutriated nanoparticle agglomerates.

A 63-mm loudspeaker, powered by a sound excitation system (Bently Navada, Series 7000) with a signal generator, is installed on the top of the fluidized bed. A precision sound pressure level meter (Brüel and Kjær, Type 2232) is used to measure the sound pressure level. The sound excitation system is capable of generating a sound wave in the fluidized bed with a sound pressure level up to 125 dB. The sound frequency from the signal generator can be adjusted from 10 to 2 MHz. The fluidization behavior is visualized with the aid of a lighting device (Illumination Technologies, Model

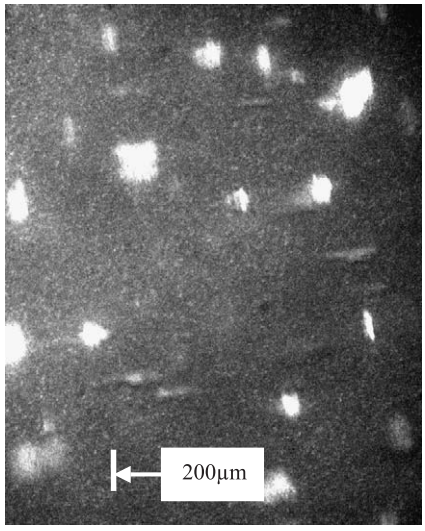
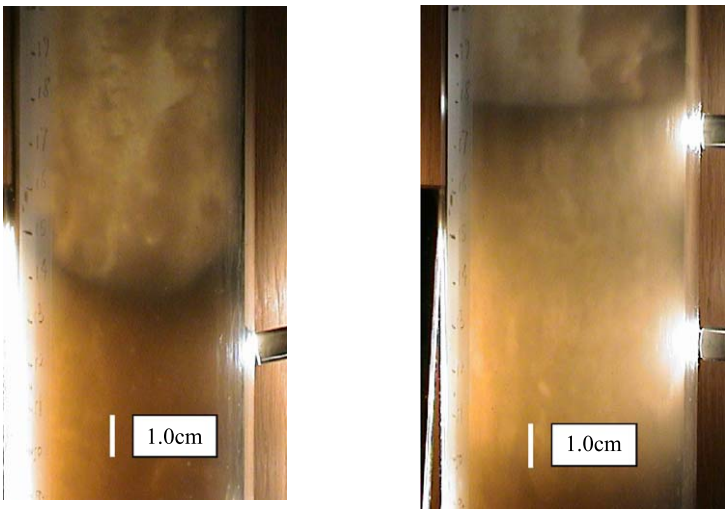


Fig. 2. Picture of agglomerates near the fluidized bed surface. (With sound f = 200Hz, 125 db, Uair = 0.1am/s).



(a) Without sound (b) With sound, 100Hz, 122dB, Uair=0.054cm/s

Fig. 3. Fluidization of SiO_2 nanoparticles.

150SX), recorded by a digital camcorder (Sony, Digital 8) and analyzed directly by a PC computer.

The nanoparticles used in this study are synthetic silicon dioxide (Degussa, R974) with a primary particle diameter of 12 nm and a primary density of 2560 kg/m^3. Before the experiments, the particles were sieved using a shaker (Octagon 2000) and a sieve of mesh no. 35 (mesh opening, about 500 μm). The sieving process serves to remove very large agglomerates, which may have been generated during packing, storage and transportation. The selection of a mesh opening of 500 μm is based on previous experimental findings that the typical size of fluidized nanoparticle agglomerates is between 100 and 400 μm. This typical size range of fluidized nanoparticle agglomerates is also validated in the current study, with the help of a laser source (Laser Physics Reliant 1000 m), a CCD camera and an image processing system. As shown in Fig. 2, agglomerate images taken near the fluidized bed surface are within the size range of 100–400 μm.

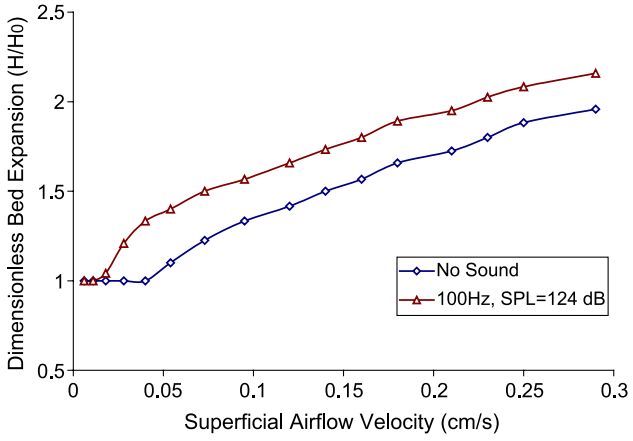


Fig. 4. Bed expansion with and without sound excitation. (H_0 is the initial bed height, which is 12.0 cm in this study).

The bulk density of sieved nanoparticle agglomerates is 33.8 kg/m^3. Due to surface treatment by the manufacturer, the nanoparticles are hydrophobic. To minimize any effect of humidity on the nanoparticle fluidization, pure nitrogen from a compressed N_2 tank is used as the fluidizing gas. The gas flow rate is measured and adjusted by a calibrated rotameter. With the aid of an inclined tube manometer, the pressure drop across the bed is measured, which excludes the pressure drop over the distributor.

3. Results and discussion

3.1. Sound assisted fluidization

Typical bed behavior of SiO_2 nanoparticle agglomerates with and without sound excitation are shown in Fig. 3. The nanoparticle agglomerates were first lifted in a slugging

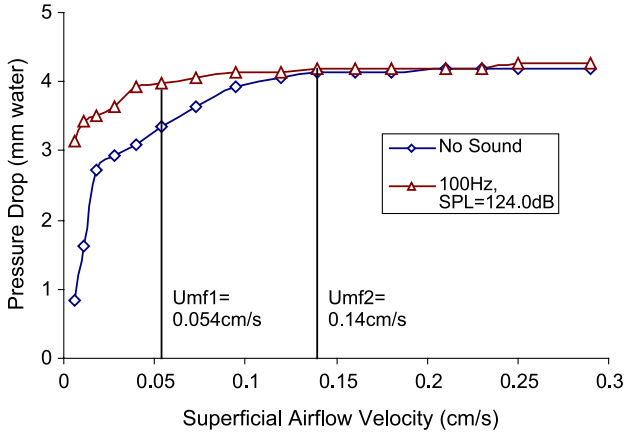


Fig. 5. Pressure drop with and without sound excitation. (U_{mf1} is the minimum fluidization velocity with sound agitation; U_{mf2} is the minimum fluidization velocity without sound agitation).

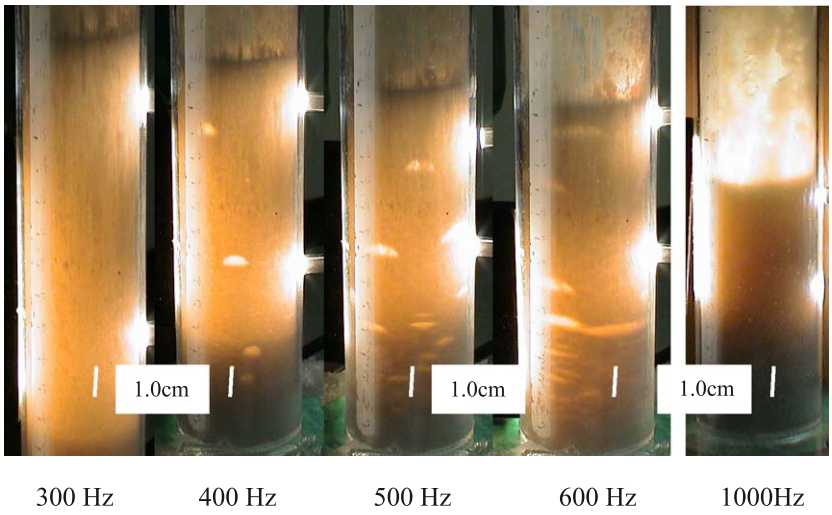


Fig. 6. Fluidization at different frequencies. (SPL=125 db, Uair=0.1cm/s).

mode and then the bed disintegrated to form stable channels. The bed only expands slightly with an uneven surface, as shown in Fig. 3(a). Once a sufficiently strong sound is applied, the bed collapses in a couple of seconds, the channels disappear, and the bed expands rapidly and uniformly until it reaches the full expansion. A homogenous fluidization state is easily established, as shown in Fig. 3(b).

Typical fluidization characteristics, including the minimum fluidization velocities, bed expansions and bed pressure drops with and without sound excitation are illustrated in Figs. 4 and 5. A substantial reduction in the minimum fluidization velocity with the aid of the sound can be observed. For DeGussa Aerosil® R974, the minimum fluidization velocity is reduced from 0.14 cm/s in the absence of sound to 0.054 cm/s with sound excitation. Here, the minimum fluidization velocity is defined as the gas superficial velocity beyond which the bed pressure drop is no longer dependent upon the gas velocity and becomes constant.

As mentioned earlier, at low gas velocities, only the slugging and channeling occur in a fluidized bed of nanoparticle agglomerates while, at sufficiently high gas velocities, the bed can be fluidized smoothly. Fluidization of nanoparticle agglomerates occurs due to the effective breakup of large agglomerate clusters by the large hydrodynamic forces at high gas velocities. With the aid of sound, the breakup of large agglomerate clusters takes place due to a combined effect of hydrodynamic forces and acoustic excitations. A more comprehensive explanation of this combined effect requires further experimental and modeling efforts.

3.2. Effects of sound frequency and sound pressure level

Fig. 6 shows a series of representative snapshots of the fluidizing bed at different sound frequencies. At a fixed sound level output (e.g., 125 dB in Fig. 6), the bed of nanoparticle agglomerates can only be fluidized in a relatively narrow band of low sound frequency from 20 to 1000 Hz. This is similar to what was observed when using sound-assisted fluidization of micron or submicron

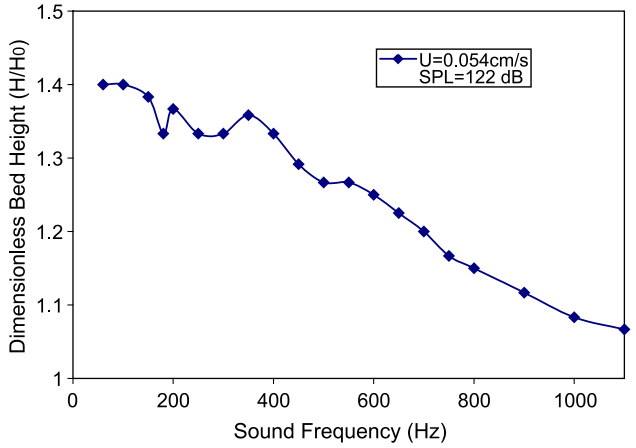


Fig. 7. Effect of sound frequency on bed expansion. (H_0 is the initial bed height, which is 12.0 cm in this study).

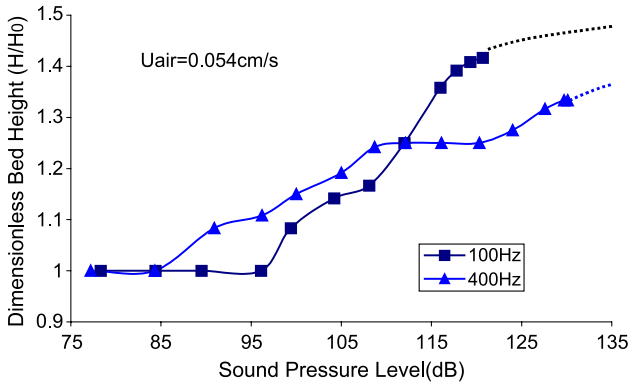


Fig. 8. Effect of sound pressure level on bed expansion. (H_0 is the initial bed height, which is 12.0 cm in this study).

particles or agglomerates. Furthermore, bubbles appear in an even narrower range, 200–600 Hz, and, as seen in Fig. 6, both the occurrence of bubbling and bubble size are strongly dependent on the sound frequency. Due to the relatively high bed voidge observed when fluidizing nanoparticle agglomerates, in the bubbling fluidization regime, the bubble size and the bubble rising velocity can be easily identified using our visualization technology. The bed expansion is also strongly dependent on the sound frequency, as seen in Fig. 7. It appears that both bed expansion and bubble formation could be linked to the wave modes of the sound in the bed, the resonance frequencies of the agglomerate clusters, and the resonance frequencies of the bed.

The effect of sound pressure level on the bed expansion is shown in Fig. 8. It is noted that below a critical value of sound pressure level (e.g., 112 dB at 100 Hz and 105 dB at 400 Hz in Fig. 8), there is no fluidization. The critical sound pressure level appears to be a function of sound frequency. The bed expansion increases monotonically as the sound pressure level increases in the range of our study. The bed expansion may be related to the balance between the sound-assisted agglomerate breakup and the sound-assisted agglomeration of the nanoparticles.

4. Conclusions

This preliminary study has shown that nanoparticle agglomerates can be easily and smoothly fluidized with the assistance of sound. Since there is a significant reduction in the minimum fluidization velocity in the presence of sound, elutriation of nanoparticle agglomerates becomes much reduced. The ability to fluidize these fumed silica nanoparticle agglomerates can only be achieved within a given range of sound frequency with a sound pressure level above a critical value. Bubbling fluidization occurs within an even smaller range of sound frequency. A larger experimental study using a variety of different nanoparticles, as well as a comprehensive mechanistic explanation of these interesting phenomena is in progress and will be reported in a subsequent paper.

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References

[1] J. Chaouki, C. Chavarie, D. Klvana, Effect of interparticle forces on the hydrodynamic behavior of fluidized aerogels, Powder Technol. 43 (1985) 117–125.
[2] S. Morooka, K. Kusakabe, A. Kobata, Y. Kato, Fluidization state of ultrafine powders, J. Chem. Eng. Jpn. 21 (1) (1988) 41–46.
[3] S. Matsuda, H. Hatano, T. Muramota, A. Tsutsumi, Particle and bubble behavior in ultrafine particle fluidization with high G, Fluidization X, Eng. Foundrym. (2001) 501–508.
[4] S. Matsuda, H. Hatano, A. Tsutsumi, Modeling for size reduction of agglomerates in nanoparticle fluidization, AIChE 2002 Annual Meeting, November 3–8, Indianapolis, Indiana, 2002, 138e.
[5] S. Matsuda, H. Hatano, A. Tsutsumi, Ultrafine particle fluidization and its application to photocatalytic Nox treatment, Chem. Eng. J. 82 (2001) 183–188.
[6] Y. Wang, F. Wei, Y. Jin, T. Luo, Bed collapse behavior of primary nanoparticles, in: M. Kwauk, J. Li, W.-C. Yang (Eds.), Proceedings of the Tenth Engineering Foundation Conference on Fluidization, United Engineering Foundation, New York, USA, Fluidization X, (2001) 477–484.
[7] Y. Wang, F. Wei, Y. Jin, T. Luo, Agglomerate particulate fluidization and E-particles, Proceedings of the Third Joint China/USA Chemical Engineering Conference (CUChE-3), 12-006, Beijing, 2000.
[8] Y. Wang, G. Gu, F. Wei, J. Wu, Fluidization and agglomerate structure of SiO_2 nanoparticles, Powder Technol. 124 (2002) 152–159.
[9] C. Nam, R. Pfeffer, R.N. Dave, S. Sundaresan, AIChE J. (2004) (in press).
[10] R. Pfeffer, R.N. Dave, personal communication (2003).
[11] C. Nam, R. Pfeffer, Aeration vibrofluidization of nanoparticles, AIChE 2002 Annual Meeting, November 3–8, Indianapolis, Indiana, 2002.
[12] R.D. Morse, Sonic energy in granular solid fluidization, Ind. Eng. Chem. 47 (6) (1955) 1170–1175.
[13] R. Chirone, L. Massimilla, S. Russo, Bubble-free fluidization of a cohesive powder in an acoustic field, Chem. Eng. Sci. 48 (1) (1993) 41–53.
[14] P. Russo, R. Chirone, L. Massimilla, S. Russo, The influence of the frequency of acoustic waves on sound-assisted fluidization of beds of fine particles, Powder Technol. 82 (1995) 219–230.
[15] R. Chirone, P. Russo, Resonant behavior of cluster–subcluster structures in sound assisted fluidization beds, Fluidization VIII (1995) 389–397.
[16] R. Chirone, L. Massimilla, Sound-assisted aeration of beds of cohesive solids, Chem. Eng. Sci. 49 (8) (1994) 1185–1194.
[17] E.K. Levy, H. Shnitzer, T. Masaki, J. Salmento, Effect of an acoustic field on bubbling in a gas fluidized bed, Powder Technol. 90 (1997) 53–57.
[18] C.A. Herrera, E.K. Levy, Bubbling characteristics of sound-assisted fluidized beds, Powder Technol. 119 (2001) 229–240.
[19] C.A. Herrera, E.K. Levy, Characteristics of acoustic standing waves in fluidized beds, AIChE J. 48 (3) (2002) 503–513.
[20] R. Chirone, L. Massimilla, S. Russo, Bubbling fluidization of a cohesive powder in an acoustic field, Fluidization VII (1992) 545–553.
[21] W. Nowak, M. Hasatani, M. Derczynski, AIChE Symp. Ser. 89 (1993) 137.
[22] J. Jung, D. Gidaspow, Fluidization on nano-size particles, AIChE Annual Meeting, Reno, NV, Nov. 6, 2001.