Enhanced Fluidization of Nanoparticles in an Oscillating Magnetic Field

Qun Yu, Rajesh N. Dave, and Chao Zhu
Dept. of Mechanical Engineering, New Jersey Institute of Technology, Newark, NJ 07102

Jose A. Quevedo and Robert Pfeffer
Dept. of Chemical Engineering, New Jersey Institute of Technology, Newark, NJ 07102

DOI 10.1002/aic.10479
Published online May 18, 2005 in Wiley InterScience (www.interscience.wiley.com).

Some experimental observations on the fluidization characteristics of nanoparticles in the form of agglomerates with magnetic assistance are presented. The nanoagglomerates consist of Degussa Aerosil® R974 fumed silica, with a primary particle size of 12 nm. An oscillating AC magnetic field is used to excite large (mm size) permanent magnetic particles mixed in with the nanoparticle agglomerates, and the fluidization behavior of the nanoagglomerates, including the fluidization regime, the minimum fluidization velocity, the bed pressure drop, and the bed expansion are investigated. It is shown that, with the aid of an oscillating magnetic field at low frequencies, the bed of nanoparticle agglomerates can be smoothly fluidized, and the minimum fluidization velocity is significantly reduced. In addition, channeling or slugging of the bed disappears and the bed expands uniformly without bubbles, and with negligible elutriation. The bed expansion and the minimum fluidization velocity depend on the mass ratio of magnetic particles to nanoparticles, and the intensity and frequency of the oscillating magnetic field. © 2005 American Institute of Chemical Engineers *AIChE J,* 51: 1971–1979, 2005
Keywords: fluidization; nanoparticles; nanoagglomerates; oscillating magnetic field; permanent magnetic particles, aggregate fragmentation

Introduction

Gas fluidization of small solid particles has been widely used in a variety of industrial applications because of its unusual capability of continuous powder handling, good mixing, large gas-solid contact area, and very high rates of heat and mass transfer. Extensive research has been done in the area of gas fluidization, and the fluidization behavior of classical powders in the size range of 30 to 1,000 μm (Geldart group A and B powders) is relatively well understood. However, the fluidization behavior of ultrafine particles, including nanoparticles, is much more complex and has received relatively little attention in the literature.

Because of their unique properties due to their very small primary particle size and very large surface area per unit mass, nanostructured materials are already being used in the manufacture of drugs, cosmetics, foods, plastics, catalysts, and energetic and bio materials. Therefore, it is necessary to develop processing technologies, which can handle large quantities of nanosized particles, for example, mixing, transporting, modifying the surface properties (coating), and downstream processing of nanoparticles to form nanocomposites. However, before processing of nanostructured materials can take place, the nanosized particles have to be well dispersed.

Gas fluidization is one of the best techniques available to disperse and process powders belonging to the Geldart group A and B classifications. Nanosized powders, however, fall under

Correspondence concerning this article should be addressed to R. N. Dave at dave@njit.edu.

the Geldart group C (<30 μm) classification, which means that fluidization is expected to be difficult due to cohesive forces, such as van der Waals forces that become more prominent as the particle size decreases.

It has been found that nanoparticles form relatively large agglomerates in order for them to fluidize,[1-14] and the minimum fluidization velocity is relatively high (about several orders of magnitude higher than the minimum fluidization velocity of primary nanoparticles,[1,2,3,4] For certain types of nanoparticles, very smooth fluidization occurs with 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.[4,5] This type of fluidization has been called agglomerate particulate fluidization (APF) by Wang et al.,[4] and has recently been comprehensively studied by Zhu et al. [6] For other types of nanoparticles, fluidization results in a very limited bed expansion, and large bubbles rise up very quickly through the bed.[6,7,8] However, even for the homogeneously fluidized nanoparticles, 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 a rotating or centrifugal fluidized bed,[15-17] where the centrifugal force acting on the agglomerates can be set much higher than gravity. We also found that the minimum fluidization velocity of nanoparticle agglomerates in a conventional fluidized bed can be significantly reduced by introducing external force excitations to the bed, such as vertical, sinusoidal vibration [5] and sound waves at relatively low frequency generated by a loudspeaker.[18] With a much lower fluidizing gas velocity, hardly any elutriation of nanoparticles was observed.

The objective of this study is to experimentally determine the fluidization behavior of a typical APF nanosized silica powder by applying an oscillating magnetic field to the nanoparticles that have been premixed with some very large magnetic particles. It is anticipated that the excitation of the magnets will provide sufficient energy to the system to overcome the interparticle forces and form stable smaller agglomerates that will fluidize smoothly at a lower minimum fluidization velocity. Another objective is to examine whether excitation by the magnets is strong enough to allow fluidization of nanopowders without any preprocessing, which often contain large, hard agglomerates.

Before describing our experiments using magnetically assisted fluidization of nanoagglomerates, it is important to review the previous work in this area. The idea of using a magnetofluidized bed was first proposed in 1960,[19] and became popular as a means of suppressing bubbles in gas fluidized beds for a variety of industrial applications. Much of the pioneering industrial work was done by Rosensweig at Exxon Corporation and was described in his excellent review article, [20] although the process never really became commercially viable. Other review articles describing the fundamental and practical development of magnetofluidized beds can be found in [21] and [22]. In fact, an entire issue of *Powder Technology* [21] was devoted to articles on magnetofluidized beds.

Generally, the particles to be fluidized were either magnetic particles or a mixture of magnetic and nonmagnetic particles, and the magnetic field was usually generated by DC current,[23-27] causing magnetic particles to form chains along the field depending on whether the magnetic field or the gas flow was applied first to the bed.[28] For example, Arnaldos et al.[24] studied the fluidization behavior of a mixture of magnetic and nonmagnetic particles of several hundred microns in size, such as sintered nickel-silica, steel-copper and steel-silica particles. The fluidization of larger particle mixtures of mm size (Geldart group D particles), such as iron-copper shot of 0.935 to 1.416 mm in dia. is described in [25] and, [26] and Lu et al.[27] studied the fluidization of very fine (Geldart group C) particle mixtures of $CaCO_3$-Fe_2O_3 in a transverse rotating magnetic field. However, in all of these studies, the magnetic particles were fluidized along with the nonmagnetic particles.

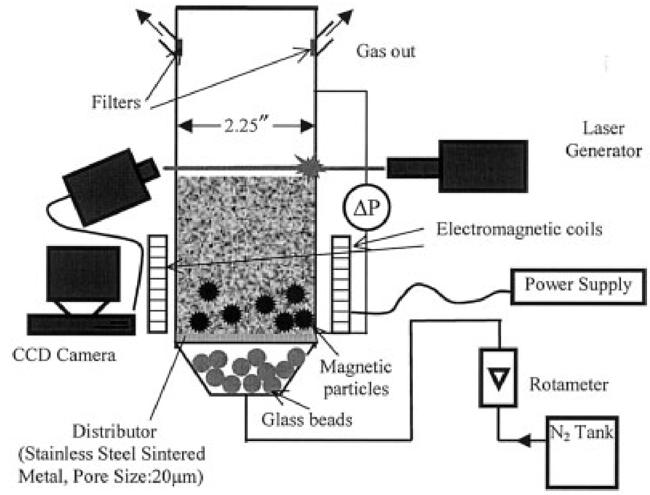


Figure 1. Experimental system.

To our knowledge, no one has studied the effect of adding large magnetic particles to a bed of nanoparticles for the purpose of disrupting the interparticle forces between the nanoparticles, so that smooth fluidization of nanoagglomerates can occur. This article presents an investigation of the fluidization behavior of nanoparticle agglomerates with the assistance of mm-sized magnetic particles excited by an oscillating magnetic field. The effects of the intensity and frequency of the oscillating magnetic field and the weight ratio of magnets to nonmagnetic nanoparticles, on important fluidization parameters, such as the minimum fluidization velocity, pressure drop across the bed, and bed expansion, will be demonstrated. It is noted that unlike traditional magnetofluidized beds, the magnetic particles used are permanent magnets, which furiously spin and create intense shear and agitations under an oscillating magnetic field.

Experimental Method

The fluidization system is shown in Figure 1. The system consists of a fluidized bed of nanoparticle agglomerates, an oscillating electromagnetic field, 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 dia. 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. To generate a

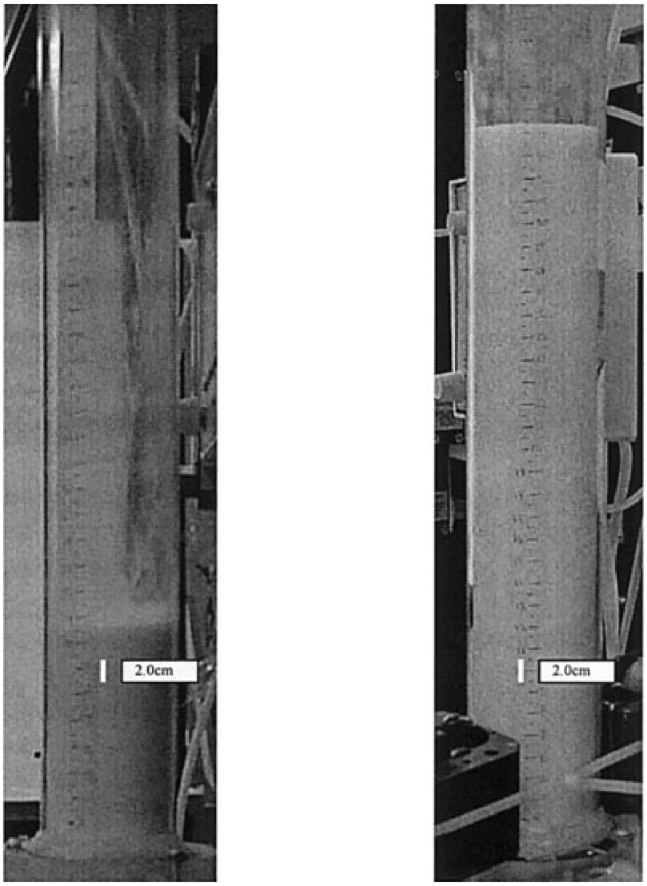


Figure 2. Fluidization of SiO_2 nanoparticles at U gas = 0.65 cm/s: (80/20 mixture).

(a) Without magnetic field; (b) with magnetic field, 140G, 60 Hz, mass ratio of magnets to nanoparticles 2:1.

uniform gas field before the distributor, glass beads of dia. between 2.5 and 3.5 mm are charged into a chamber placed below the distributor and above the gas inlet to form a packed bed about 100 mm high. An ultrafine mesh filter is located at the gas outlet to filter out any elutriated nanoparticle agglomerates. The fluidization behavior is visualized with the aid of a lighting device (Illumination Technologies, Model 150SX), and recorded by a digital camcorder (Sony, Digital 8).

The magnetic particles are barium ferrite ($BaO\text{-}6Fe_2O_3$) coated with polyurethane (supplied by Aveka, U.S.A.), about 1.0 – 3.0 mm in size. These are permanent magnetic particles, which are recharged by contacting them with a strong permanent magnet before each experiment and are then added to the bed of nanoparticles at a prescribed mass ratio. The shafts of two 1/20 HP electric motors (Dayton 5M064B) are removed and the electromagnetic coils are placed opposite one another around the lower part of the vertical transparent column by mounting them on the acrylic plate which holds the distributor, as shown in Figure 2b. The coils are driven by an alternating current generated by a power supply and can generate an oscillating magnetic field with an intensity up to 140 Gauss at the center of the coil. The power supply (Triathlon Precision AC Source) can supply AC current with adjustable frequency and voltage. A strong cooling fan (Comair Rotron TNE2A) is used to prevent the coils from overheating.

Fumed SiO_2 nanoparticles (Degussa Aerosil® R974) with a primary particle size of 12 nm and a bulk density of about 30 kg/m^3 are used in this study. Due to surface treatment by the manufacturer, the nanoparticles are hydrophobic. Before the experiments, the particles are sieved using a shaker (Octagon 2000) and a 35-mesh sieve opening (about 500 μm). The sieving process serves to separate 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 to 400 μm [1, 2, 6, 7, 12]. The size range of the fluidized nanoparticle agglomerates is measured by analyzing digital images of the fluidized agglomerates with the help of a laser source (Laser Physics Reliant 1000m), a CCD camera (LaVision FlowMaster 3S), and an image processing system (Dual Xeon CPU).

We designate the smaller nanoagglomerates, which pass through the openings of the 500 μm sieve as "soft" and the larger agglomerates, from about 500 μm to more than 10 mm as "hard". These two different sized agglomerates and a "mixture" consisting of 80% soft agglomerates and 20% hard agglomerates by weight (80/20), were selected to conduct the fluidization experiments.

To minimize any effect of humidity on the fluidization experiments, pure nitrogen from a compressed N_2 tank is used as the fluidizing gas. The gas flow rate is measured and adjusted by two calibrated rotameters (Gilmont) with a combined flow rate range of up to 51.0 liters per min. The pressure drop across the bed is measured with a differential pressure transmitter (Cole-Parmer) with a measurement range of up to 1.0 in. of water; the lower pressure tap is placed slightly above the distributor (approximately 3 mm), so that it is not necessary to measure the pressure drop across the distributor. A Gaussmeter (Walker Scientific, Inc. MG-3A) with a range of from 1 to 10^4 G is used to measure the intensity of the oscillating magnetic field, which is measured at the center point between the coils in the empty column (before charging the nanoparticles into the bed).

Results and Discussion

Magnetic assisted fluidization

We have found that, even when using the same nanoparticles, if we select agglomerates of different sizes, the bed will show very different fluidization behavior. For example, the soft R974 agglomerates fluidize smoothly with large bed expansion (APF) at a low minimum fluidization velocity of 0.23 cm/s. Here, we define the minimum fluidization velocity as the gas superficial velocity beyond which the bed pressure drop is no longer dependent on the gas velocity and becomes constant, and a relatively large bed expansion (typically 2 or more times the initial bed height) occurs. The mixture consisting of 80% soft agglomerates, and 20% hard agglomerates (80/20) also behaves as APF, but the minimum fluidization velocity is much higher (5.67 cm/s) than that of the soft agglomerates. However, the hard R974 agglomerates do not fluidize at all, even at a gas velocity as high as 13.2 cm/s. At this high gas velocity, significant particle elutriation was observed, and the fluidization experiment had to be interrupted to avoid large losses of nanoparticles.

Typical fluidization behavior of the 80/20 mixture of SiO_2

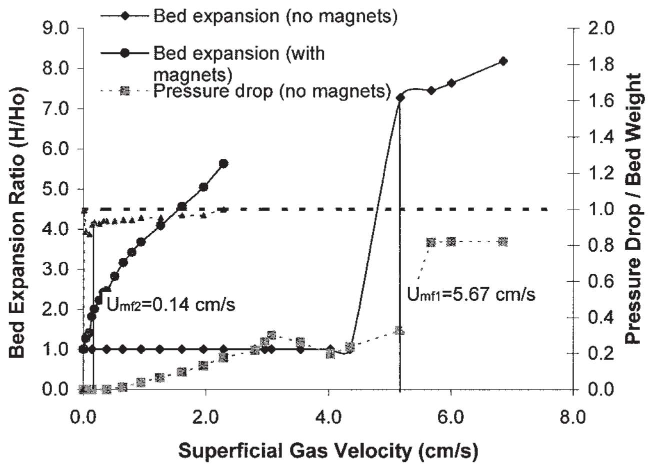


Figure 3. Bed expansion ratio and pressure drop for 80/20 mixture with and without magnetic excitation.

(Solid lines are the bed expansion ratios, and dashed lines are the pressure drops.)
(Magnetic field intensity 140G at the center of the field, mass ratio of magnets to nanoparticles 2:1, AC frequency 60 Hz.) Umf1: minimum fluidization velocity without magnetic excitation; Umf2: minimum fluidization velocity with magnetic excitation.

nanoparticle agglomerates with and without the external oscillating magnetic excitation is shown in Figure 2. Without the external oscillating magnetic excitation, at a superficial gas velocity of 0.65 cm/s (Figure 2a), the nanoparticle agglomerates are first lifted as a plug and then the plug disintegrates to form stable channels through which the gas passes; the bed expands slightly with an uneven surface and the pressure drop is much less than the bed weight, indicating that the nanoagglomerate bed is not fluidized.

However, if a sufficiently strong oscillating magnetic field is applied, the magnetic particles are set in motion (translation and rotation) and the nanoparticle agglomerates are fragmented into smaller agglomerates because of collisions with the magnets, the vessel wall, and the distributor. After a few minutes, the channels disappear, and the bed begins to expand slowly and uniformly until it reaches its full expansion, of up to five times the initial bed height, and at the same time, the pressure drop reading is very close to the weight of the bed, indicating fluidization of the entire bed. A homogeneous fluidization state is established, as shown in Figure 2b, and the surface is very smooth and even. After the experiment, the powder is poured out, and from visual observation, most of the original large hard agglomerates are gone and the average agglomerate size appears very much smaller.

The pressure drop normalized with the bed weight per unit area and the bed expansion ratio as a function of superficial gas velocity through the bed is shown in Figure 3, with and without magnetic excitation. It is clear from the figure that the magnetic excitation causes the bed to expand almost immediately as the velocity is increased and the bed fluidizes at a velocity more than one order of magnitude lower than that without magnetic assistance.

After separation from the magnetic particles, the nanoparticle agglomerates are recharged back into the column, and a second fluidization experiment without magnetic assistance is conducted using these agglomerates. Figure 4 is a comparison of the fluidization characteristics of the 80/20 mixture, before and after magnetic processing. A significant reduction in the minimum fluidization velocity from 5.67 cm/s to 1.25 cm/s is observed, indicating that previous fluidization with magnetic assistance causes the agglomerates to be fragmented into smaller ones and the average agglomerates size is reduced. However, the minimum fluidization velocity of these smaller agglomerates is still about an order of magnitude larger than the minimum fluidization velocity observed when the magnetic assistance is turned on.

The fluidization behavior of the soft agglomerates is shown in Figure 5. These much smaller agglomerates fluidize well with and without magnetic excitation. In both cases, the minimum fluidization velocities appear to be quite close to each other, but at higher gas velocities (above minimum fluidization velocity) the bed expansion with magnetic assistance is higher than that without magnetic assistance. It should also be noted that the ratio of the measured pressure drop to the weight of the bed per unit area is below unity for magnetic assisted fluidization. This may mean that some of the nanoagglomerates are not participating in the fluidization and may be sticking to the magnets.

Figure 6 shows the typical fluidization behavior (pressure drop and bed expansion) of hard SiO_2 nanoparticle agglomerates (R974) with and without magnetic excitation. The size of the hard agglomerates is in a wide range, from 0.5 mm to about 10 mm. Without the magnetic excitation, even at a superficial gas velocity as high as 13.2 cm/s, the hard agglomerates could not be fully fluidized. Visual observation reveals that the smaller hard agglomerates are in motion at the top of the bed, but the larger agglomerates remain the bottom of the bed,

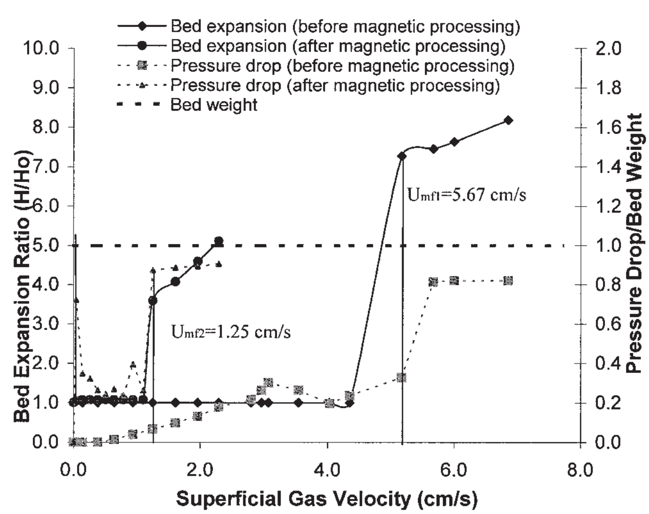


Figure 4. Bed expansion ratio and pressure drop for conventional fluidization of 80/20 mixture before and after magnetic processing.

(Solid lines are the bed expansion ratios, and dashed lines are the pressure drops.)
(Magnetic field intensity 140G at the center of the field, mass ratio of magnets to nanoparticles 2:1, AC frequency 60 Hz.) Umf1: minimum fluidization velocity before magnetic "fragmentation" processing; Umf2: minimum fluidization velocity after magnetic "fragmentation" processing.

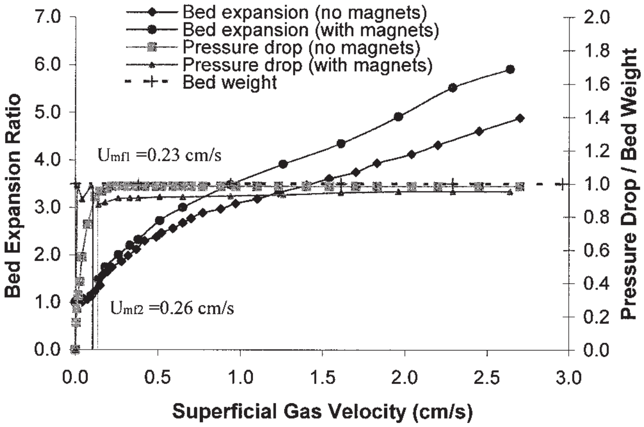
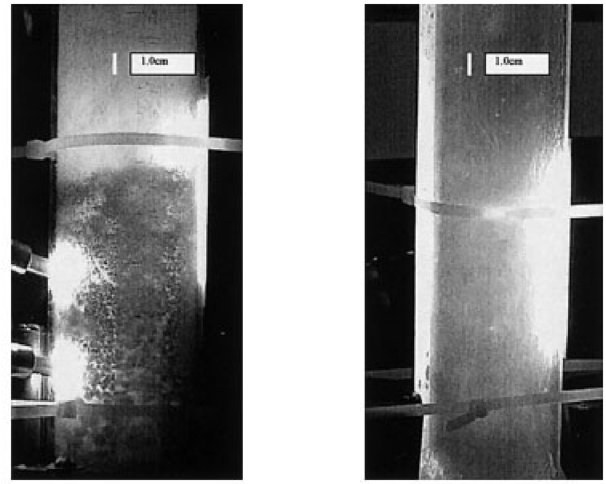


Figure 5. Bed expansion ratio and pressure drop for soft agglomerates with and without magnetic excitation.

(Solid lines are the bed expansion ratios, and dashed lines are the pressure drops.)
(Magnetic field intensity 140G at the center of the field, mass ratio of magnets to nanoparticles 2:1, AC frequency 60 Hz.)
Umf1: minimum fluidization velocity without magnetic excitation; Umf2: minimum fluidization velocity with magnetic excitation.



(a) Without magnetic field (b) With magnetic field

Ugas=13.15 cm/s Ugas=0.94 cm/s

Figure 7. Fluidization of hard agglomerates.

(140G, 60Hz, mass ratio of magnets to nanoparticles 2:1.)

causing the gas to flow in large channels between them. The bed shows almost no expansion (see Figure 7a), and the pressure drop is much less than the bed weight, indicating that the bed is not fluidized.

After turning on the external magnetic field, however, the large agglomerates become smaller and smaller due to fragmentation (disruption of interparticle forces) caused by collisions with the magnetic particles, and these smaller agglomerates participate in the circulation of the bed. After a few minutes, even at the relatively low gas velocity of 0.94 cm/s, all of the large agglomerates disappear, and the bed expands slowly and uniformly until it reaches the full expansion, (Figure 7b), while the pressure drop reading is very close to the weight of the bed, indicating that the entire bed is fluidized.

The fragmentation caused by the magnetic processing is so obvious that the reduction in size of the hard agglomerates could be seen by inspection after the magnetic field and air flow were shut down. Upon removing the magnetic particles, the nanoparticle agglomerates are recharged back into the chamber and a conventional fluidization experiment (no magnetic assistance) is performed. Figure 8 is a comparison of the fluidization characteristics between the powder before and after the magnetic assisted fluidization (fragmentation) process. A very large reduction in the minimum fluidization velocity (U_{mf}) from larger than 13.2 cm/s to 2.29 cm/s indicates that the average agglomerates size has been significantly reduced.

The U_{mf} for the hard agglomerates after magnetic processing is 2.29 cm/s, which is larger than the U_{mf} of 1.25 cm/s for the 80/20 mixture, and also much larger than the U_{mf} of 0.23 cm/s for the soft agglomerates. This indicates that the average size of hard agglomerates, and of the mixture after the fragmentation process is still larger than that of the soft agglomerates. Hence, in order to only investigate the effect of magnetic excitation, such as magnet to nanoparticle mass ratios, AC frequencies, and different magnetic field intensity, and to minimize the influence of nonuniformity of the initial agglomerate size distribution, the soft agglomerates are a good choice to conduct the comparison experiments.

At low gas velocities, conventional fluidization (no magnetic assistance) of soft agglomerates or of the 80/20 agglomerate mixture, produces only slugging and channeling, while at sufficiently high-gas velocities, the bed can be fluidized smoothly. If we continue to increase the gas velocity above a certain

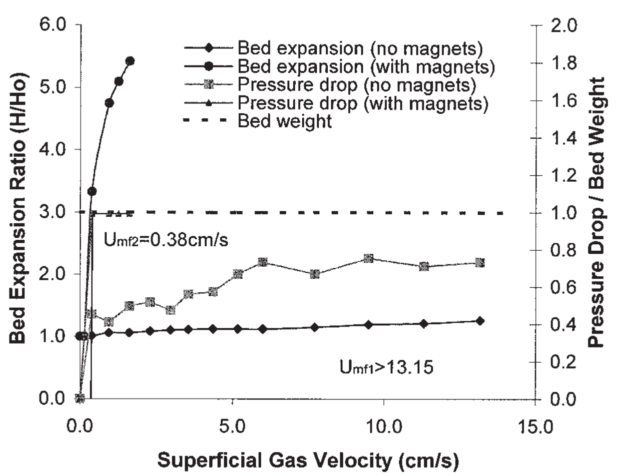


Figure 6. Bed expansion ratio and pressure drop for hard agglomerates with and without magnetic excitation.

(Solid lines are the bed expansion ratios, and dashed lines are the pressure drops.)
(Magnetic field intensity 140G at the center of the field, mass ratio of magnets to nanoparticles 2:1, AC frequency 60 Hz)
Umf1: minimum fluidization velocity without magnetic excitation; Umf2: minimum fluidization velocity with magnetic excitation.

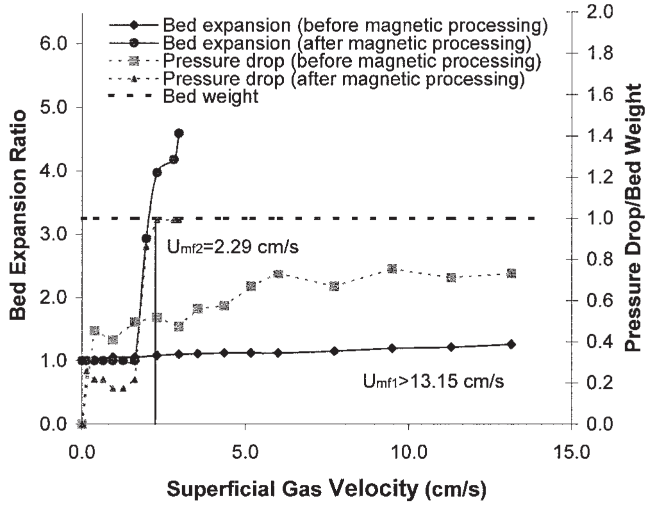


Figure 8. Bed expansion ratio and pressure drop for conventional fluidization of hard agglomerates before and after magnetic processing.

(Solid lines are the bed expansion ratios, and dashed lines are the pressure drops.)
(Magnetic field intensity 140G at the center of the field, mass ratio of magnets to nanoparticles 2:1, AC frequency 60 Hz)
Umf1: minimum fluidization velocity before magnetic "fragmentation" processing; Umf2: minimum fluidization velocity after magnetic "fragmentation" processing.

level, bubbles can be observed in the fluidized bed. Fluidization of nanoparticle agglomerates occurs due to the disruption of interparticle forces by the large hydrodynamic forces generated at high gas velocities. However, for conventional fluidization of hard agglomerates, even at a very high-gas velocity, the bed could not be fully fluidized.

The mechanism of fluidization with the assistance of an oscillating magnetic field is much more complicated. The excited magnets will enhance nanoparticle fluidization in two possible ways: fragmentation of large agglomerates into smaller ones, and transferring kinetic energy generated by the oscillating magnetic excitation to the nanoparticle agglomerates due to collisions to disrupt the large interparticle forces between them. A more comprehensive explanation of this combined effect requires further experimental and modeling efforts.

Table 1 presents a summary of the minimum fluidization velocities for the soft, hard and 80/20 agglomerate mixture. For the soft agglomerates magnetic excitation has little effect, but it produces a definite improvement in fluidization behavior for the 80/20 mixture. Even for the hard agglomerates, magnetic excitation changes the fluidization characteristics significantly, from no fluidization to smooth, bubbleless, agglomerate particulate fluidization (APF), with a very large bed expansion up to five times of the initial bed height. The minimum fluidization velocity is also significantly reduced from higher than 13.2 cm/s to 0.38 cm/s. Without magnetic excitation, at a gas velocity of 13.2 cm/s or higher, extremely strong elutriation could be observed, while with magnetic excitation, at the low-gas velocity of 0.38 cm/s, elutriation was negligible. The substantial reduction in the minimum fluidization velocity resulting in smooth and bubbleless fluidization with little elutriation should be very beneficial to industrial applications where good mixing and high rates of heat and mass transfer with little gas bypassing are required.

In-situ agglomerates size measurement

Agglomerate size is one of the key variables, which significantly influences the fluidization characteristics. Models to predict agglomerate size have been proposed for cohesive fine particles [29] and for nanoparticles.[1,5,6,9,14] *In vitro* measurement of agglomerate sizes by use of SEM and/or particle counters can result in significant errors if the agglomerates are fragile and break easily when they are removed from the fluidized bed and during sample preparation. In a previous article,[6] we proposed an *in situ* method to measure agglomerate size on the fluidized bed surface by use of an optical system consisting of a laser source, a CCD camera, and an image processing system.

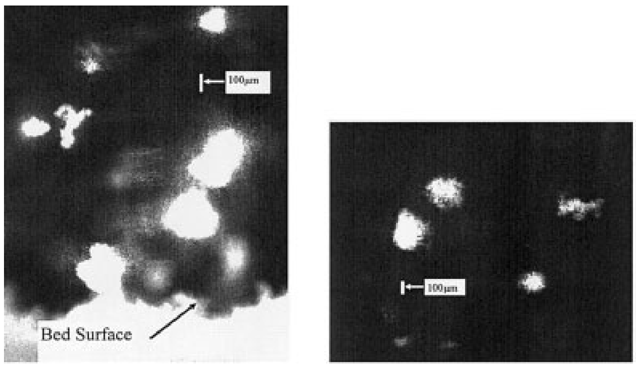
Figure 9 shows typical images of the agglomerates on the fluidized bed surface, and Figure 10 shows the agglomerate size distributions, with and without magnetic assistance. The results are given in Table 2, which shows that the mean size of R974 agglomerates is decreased from 315 μm to 196 μm by the magnetic processing. However, because of the relatively small number of images taken (less than 200), and the relatively large standard deviations obtained, the error in the mean size of the agglomerates may be appreciable, although the decrease in agglomerate size due to magnetic processing is clear. Therefore, following the methodology developed by our group in a previous article,[6] we also calculated the mean agglomerate size based on experimental measurements of bed expansion and superficial gas velocity for both cases with and without magnetic excitation. These calculated results, also listed in Table 2, show that the mean size of R974 silica nanoparticle agglomerates decreases from 211 μm to 95 μm after magnetic processing. Both the optically measured and the calculated agglomerate sizes indicate that the mean agglomerate size decreases by roughly 100 μm during the magnetic processing.

Bed expansion and collapse as a function of time

According to experimental observation, when the magnetic excitation is turned on the fluidization behavior of the nanoparticle bed does not change immediately, and it will take

Table 1. Minimum Fluidization Velocities for Soft Agglomerates, Hard Agglomerates and 80/20 Mixture

Experimental Conditions	Soft Agglomerates Umf (cm/s)	Hard Agglomerates Umf (cm/s)	80/20 Mixture Umf (cm/s)
Before processing, conventional fluidization	0.23	>13.2	5.67
During magnetic processing, magnetic assisted fluidization	0.26	0.38	0.14
After magnetic processing (magnets removed from the processed nano-powder), conventional fluidization	0.23	2.29	1.25



(a) Without magnetic field (b) With magnetic field, 140G, 60Hz, mass ratio of magnets to nanoparticles 2:1

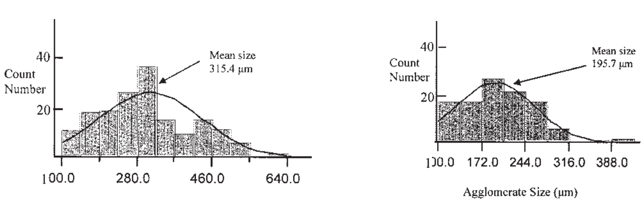
Figure 9. Photographic images of agglomerates near the fluidized bed surface at Ugas = 0.5 cm/s.

(Soft agglomerates); (a) Without magnetic field; (b) with magnetic field, 140G, 60Hz, mass ratio of magnets to nanoparticles 2:1

several minutes for the bed to begin expanding, and not reach full expansion until after about 5 to 15 min. The bed expansion as a function of time for R974 silica at different gas velocities is shown in Figure 11a; the higher the velocity, the quicker the bed expansion. Similarly, when turning off the magnetic excitation, it also takes a short period of time, typically 10 – 30 s, for the bed to begin to collapse, and the collapse will last from 1 to 3 min before reverting back to a fixed bed with uneven surface. The bed collapse as a function of time is shown in Figure 11b; the higher the gas velocity, the longer it will take for the bed to collapse.

Effects of mass ratio of magnets to nanoparticles

Fluidization experiments with magnetic assistance were conducted using the soft agglomerates for four different mass ratios of magnets to nanoparticles, varying from 1:4 to 2:1. Table 3 presents the values of U_{mf} and the bed expansion ratios at two different gas superficial velocities that were observed for these four cases. The table shows that the minimum fluidization velocity and bed expansion depends on the magnet to nanoparticle mass ratio, with U_{mf} decreasing from 1.61 cm/s to 0.26



(a) Without magnetic field (b) With magnetic field, 140G, 60Hz, mass ratio of magnets to nanoparticles 2:1

Figure 10. Typical agglomerate size distribution.

(*In situ* optical measurements on the fluidized bed surface, soft agglomerates): (a) Without magnetic field; (b) with magnetic field, 140G, 60Hz, mass ratio of magnets to nanoparticles 2:1.

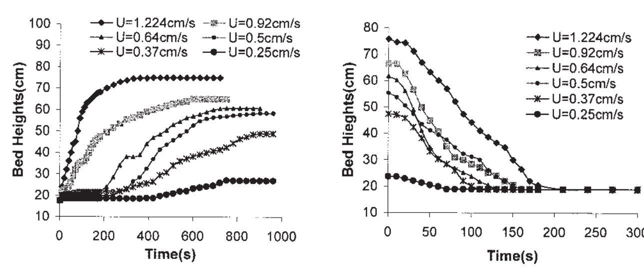
Table 2. Comparison of Agglomerate Sizes from Optical Experimental Measurements and Calculation Results Using the Prediction Methodology Developed in [13]

Magnetic Excitation	Measured Agglomerate Mean Size (μm)	Number of Samples	Standard Deviation (μm)	Calculated Results
No	315	193	123	211
Yes	196	172	64.9	95

cm/s as the mass ratio increases from 1:4 to 2:1. This indicates that adding more magnetic particles to the bed will result in more kinetic energy transported from the magnets to the nanoagglomerates, causing more fragmentation and easier fluidization. The table also shows that there is little benefit in increasing the ratio of magnets to nanoparticles above 1:1. It should also be noted that the minimum fluidization velocities for low mass ratios of magnets to nanoagglomerates are actually higher than what we observed for the nanoagglomerates without any magnetic assistance. This is probably due to the additional drag of the gas on the magnetic particles.

Effects of intensity of the oscillating magnetic field

Table 4 presents the values of U_{mf} and bed expansion ratio at a fixed superficial gas velocity for three different magnetic field intensities when fluidizing soft nanoagglomerates, keeping the ratio of magnets to nanoparticles at 2:1. We selected the center point of the column around which the 2 coils are placed as the reference point for measuring the intensity of the magnetic field and observed that when using a magnetic field intensity of less than 80G, the bed could not be fluidized. Hence, three different intensities 100, 120, and 140G are selected to conduct the fluidization experiments. Table 4 shows that the minimum fluidization velocity is a strong function of the magnetic field intensity, and U_{mf} decreases rapidly as the intensity of the magnetic field increases, indicating better fluidization. The values of the bed expansion are quite close to one another, but still, they show the expected trend that the bed will expand more in a stronger magnetic field.



(a) Bed expansion. (b) Bed collapse.

Figure 11. Bed expansion and collapse for soft agglomerates with magnetic excitation as a function of time.

(Magnetic field intensity 140G at the center of the field, mass ratio of magnets to nanoparticles 2:1, AC frequency 60 Hz.); (a) Bed expansion; (b) bed collapse

Table 3. Minimum Fluidization Velocities and Bed Expansion Ratios for Soft Agglomerates with Different Mass Ratios of Magnets to Nanoparticles

Mass Ratio (Magnets to Nanoparticles)	Umf (cm/s)	Bed Expansion Ratios at Ugas = 1.26 cm/s	Bed Expansion Ratios at Ugas = 1.96 cm/s
1:4	1.61	1.35	3.82
1:2	0.51	3.57	4.43
1:1	0.26	3.35	4.09
2:1	0.26	3.91	4.91

Magnetic field intensity 140G at the center of the field, AC frequency 60 Hz.

Table 5. Minimum Fluidization Velocities and Bed Expansion Ratios for Soft Agglomerates with Magnetic Assistance at Different Frequencies

Frequency of Magnetic Field (Hz)	Umf (cm/s)	Bed Expansion Ratios at Ugas = 2.29 cm/s
45	0.65	5.26
60	0.51	5.30
80	2.64	2.17 (Ugas = 2.64 cm/s)

Mass ratio of magnets to nanoparticles 2:1, magnetic field intensity 120 G at the center of the field.

Effects of frequency of the oscillating magnetic field

Table 5 presents the values of U_{mf} and bed expansion ratio at a fixed superficial gas velocity for three different frequencies of AC power, keeping the mass ratio of magnets to nanoparticles at 2:1 and the magnetic field intensity at 120G. The table shows that the frequency of the magnetic field can significantly affect the minimum fluidization velocity. At the lower frequencies, 45 Hz and 60 Hz, the beds show similar fluidization behavior, and can be fluidized easily at a U_{mf} of 0.65 cm/s and 0.51 cm/s, respectively. However, at higher frequency, 80 Hz, the bed is difficult to fluidize, U_{mf} is as high as 2.64 cm/s, and the bed expansion is much smaller than that at the lower frequencies. At a frequency higher than 90 Hz, the bed could not be fluidized at all.[5,18]

Concluding Remarks

This study has shown that silica nanoparticle agglomerates can be easily and smoothly fluidized with the assistance of magnetic particles in an oscillating magnetic field. Due to a significant reduction in the minimum fluidization velocity with magnetic assistance, both elutriation of nanoparticle agglomerates and gas bypass in the form of bubbles is greatly reduced. With magnetic excitation, hard (larger than 500 μm) agglomerates change their fluidization pattern from no fluidization to agglomerate particulate fluidization (APF) with large bed expansion. The minimum fluidization velocity of an 80% soft (smaller than 500 μm) and 20% hard agglomerate (80/20) mixture can also be significantly reduced, resulting in easier and more uniform fluidization, indicating that this approach can be used for as received powders, straight out of the bag, without any preprocessing, and, hence, is very useful for practical applications.

From *in situ* agglomerate size measurements on the surface of the fluidized bed and calculations using a predictive model based on experimental data, it is found that magnetic excitation will result in fragmentation of the agglomerates, so that the mean agglomerate size is significantly reduced. The ability to fluidize these fumed silica nanoparticle agglomerates depends on the mass ratio of magnets to nanoparticles, the intensity of the magnetic field, and the frequency of the magnetic field. A more comprehensive experimental study using a variety of different nanoparticles, as well as a mechanistic explanation of these interesting phenomena is in progress.

Table 4. Minimum Fluidization Velocities and Bed Expansion Ratios for Soft Agglomerates with Different Intensities of Magnetic Field

Intensity of Magnetic Field (Gauss)	Umf (cm/s)	Bed Expansion Ratios at Ugas = 2.29 cm/s
100	2.29	5.13
120	0.51	5.30
140	0.26	5.52

Mass ratio of magnets to nanoparticles 2:1, AC frequency 60 Hz.

Acknowledgments

We thank the National Science Foundation for financial support through Grant # 0210400, NIRT - Collaborative Research: Experimental and Computational Investigations of Fluid Interactions/Transport in Nanodomains and Around Nanoparticles. Partial support from the New Jersey Commission on Science and Technology (#01-2042-007-24) is also acknowledged. We would also like to express our appreciation to Dr. Herbert Riemenschneider and Mr. Jonah Klein of Degussa-Huls for supplying us with nanoparticles, Dr. Guangliang Liu for helping in the setting up the experiments, Mr. Yueyang Shen for helping in the agglomerate size measurements and Dr. Durga Misra for helping us with the magnetic field measurements.

Literature Cited

1. Chaouki J, Chavarie C, Klvana D. Effect of interparticle forces on the hydrodynamic behavior of fluidized aerogels. *Powder Technol*. 1985; 43:117-125.
2. Morooka S, Kusakabe K, Kobata A, Kato Y. Fluidization state of ultrafine powders. *J of Chem Eng of Japan*. 1988;2141-46.
3. Wang Y, Wei F, Jin Y, Luo T. Agglomerate particulate fluidization and E-particles, Proceedings of the Third Joint China/USA Chemical Engineering Conference (CUChE-3), 12-006, Beijing, 2000.
4. Wang Y, Gu G, Wei F, Wu J. Fluidization and agglomerate structure of SiO_2 nanoparticles. *Powder Technol*. 2002;124:152-159.
5. Nam C, Pfeffer R, Dave RN, Sundaresan S. Aerated vibrofluidization of silica nanoparticles. *AIChE J*. 2004;50:1776-1785.
6. Zhu C, Yu Q, Pfeffer R, Dave RN. Gas Fluidization Characteristics of Nanoparticle Agglomerates. *AIChE J*. 2005;51:426–439.
7. Wang Z, Kwauk M, Li H. Fluidization of fine particles. *Chem Eng Sci*. 1998;55377-395.
8. Pacek AW, Nienow AW. Fluidisation of fine and very dense hard metal powders. *Powder Technol*. 1990;60;145-158.
9. Zhou T, Li H. Estimation of agglomerate size of cohesive particles during fluidization. *Powder Technol*. 1999;101:57-62.
10. Jung J and Gidaspow D. Fluidization of nano-size particles. *J of Nanoparticle Research*. 2002;4:483-497.
11. Li H, Hong R, Wang Z. Fluidizing ultrafine powders with circulating fluidized bed. *Chem Eng Sci*. 1999;54:5609-5615.
12. Wang Y, Wei F, Yu H, Gu G. The large-scale production of carbon nanotubes in a nano-agglomerate fluidized-bed reactor. *Chem Phys Letts*. 2002;364:568-572.
13. Matsuda S, Hatano H, Tsutsumi A. Ultrafine particle fluidization and its application to photocatalytic NO_x treatment. *Chem Eng J*. 2001;82: 183-188.
14. Zhou T, Li H. Force balance modeling for agglomerating fluidization of cohesive particles. *Powder Technol*. 2002;111:60-65.
15. Matsuda S, Hatano H, Muramota T Tsutsumi A. Particle and bubble

behavior in ultrafine particle fluidization with high G. *Fluidization X Eng Found*. 2001;501-508.
16. Matsuda S, Hatano H, Tsutsumi A. Modeling for size reduction of agglomerates in nanoparticle fluidization. *AIChE 2002 Annual Meeting*., Indianapolis, Indiana, November 3-8; 2002:138e.
17. Quevedo JA. Fluidization of agglomerates of nanoparticles under different force fields, Master's Thesis, NJIT, 2004.
18. Zhu C, Liu G, Yu Q, Pfeffer R, Dave RN, Nam. Sound assisted fluidization of nanoparticle agglomerates. *Powder Technol*. 2004;141: 119-123.
19. Filippov MV. The effect of a magnetic field on a ferromagnetic particle suspension bed. *Prik Magnit Lat SSR*. 1960;12:215.
20. Rosensweig RE. Process concepts using field stabilized two-phase flow. *J of Electrostatics*. 1995;34:163-187.
21. Liu YA, Hamby RK, Colberg RD. Fundamental and practical developments of magnetofluidized bed: *A review*. *Powder Technol*. 1991; 64:3-41.
22. Saxena SC, Ganzha VL, Rahman SH, Dolidovich AF. Heat Transfer and relevant Characteristics of magnetofluidized bed. *Adv Heat Transfer*.1994;25:151.
23. Ganzha VL, Saxena SC. Heat-transfer characteristics of magnetofluidized beds of pure and admixtures of magnetic and nonmagnetic particles. *Int J of Heat Mass Transfer*. 1998;41:209-218.
24. Arnaldos J, Casal J, Lucas A, Puigjamer L. Magnetically stabilized fluidization: modeling and application to mixtures. *Powder Technol*. 1985;44:57-62.
25. Wu WY, Navada A, Saxena SC. Hydrodynamic characteristics of a magnetically stabilized air fluidized bed of an admixture of magnetic and non-magnetic particles. Powder Technology. 1997;90:39-46.
26. Wu WY, Smith KL, Saxena SC. Rheology of a magnetically stabilized bed consisting of mixtures of magnetic and non-magnetic particles. *Powder Technol*.1997,91,181-187.
27. Lu X, Li H. Fluidization of $CaCo_3$ and Fe_2O_3 particle mixtures in a transverse rotating magnetic field. *Powder Technol*. 2000;107:66-78.
28. Rosensweig RE, Jerauld GR, Zahn M. *Structure of magnetically stabilized fluidized solids*. :Brulin O, T RK,.Hsieh, Eds., Continum models of discrete systems. Vol 4. North-Holland, Amsterdam; 1981: 137-144.
29. Iwadate Y, Horio M., Prediction of agglomerate sizes in bubbling fluidized beds of group C powders. *Powder Technol*. 1998;100:223.

Manuscript received Apr. 14, 2004, and revision received Dec. 7, 2004.