De-agglomeration of nanoparticles in a jet impactor-assisted fluidized bed

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ABSTRACT

Nanoparticles in agglomerated state lose their outstanding properties; hence, it is essential to break them up prior to use and prevent their re-agglomeration. Even though there are several dry techniques to disperse nanopowders, none of them have been able to produce truly nanoscale aerosols so far. Here, we study de-agglomeration of dry silica nanopowder via a jet impactor-assisted fluidized bed (JIAFB). The particle size distribution of fragmented powders was characterized by in-line scanning mobility particle spectrometry (SMPS) and offline transmission electron microscopy (TEM). In order to ascertain the jet length and that the kinetic energy of particles is sufficient for de-agglomeration, a CFD simulation was carried out. Both SMPS and TEM measurements imply that at a certain fluidization velocity, increasing the jet velocity shifts the particle size distribution towards smaller diameters, and at higher velocities the mode value reduced from 113–130 to 55–60 nm. However, the geometric standard deviation or degree of polydispersity rises from 1.5 to 2.0 by increasing the jet velocity up to 197 m/s, as it will increase the total superficial velocity and consequently entrainment of larger particles from the bed. In addition, the TEM results indicate that the range of individual particle sizes in the supplied nanopowder is wide; hence, increasing the geometric standard deviation can be an indicator of a higher level of agglomerate dispersion.

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1. Introduction

Nanoparticles, due to their high surface area-to-volume ratio and free atoms on their surfaces, have a tendency to assemble together as well as absorb a wide range of molecules such as water, oxygen, etc. In other words, these features will lead the particles to have high surface energy, become unstable and be very cohesive. Therefore, individual nanoparticles, in order to reach a lower energy state, attract each other and form assemblages under the influence of some external and internal interparticle forces such as van der Waals, electrostatic, and capillary forces [1]. These friable and readily dispersed assemblages of particles are called agglomerates or "soft" agglomerates and can be formed during production, transportation or storage as a result of Brownian motion, collisions, and pressure arising from stacking. In this form, particles lose the extraordinary surface-driven properties they had as individual nanoparticles. In order to take advantage of their "nanoproperties", it is necessary to break up the agglomerates and reduce their high surface energy, or "passivate" them, before use. The ability to produce bulk quantities of highly dispersed nanoparticles is a significant limitation of nanotechnology [2].

De-agglomeration of nanopowder can be performed in the gas [3–11] or liquid phase [12–17], and a variety of theoretical and experimental studies have been conducted (Table 1). Various techniques have been developed to disperse nanoparticles in fluids, typically through use of mechanical and acoustic energy. Ultrasonication is a well-known technique to disperse nanoparticles homogeneously in suspensions using acoustic energy. Through acoustic cavitation and streaming, the formation, growth and implosion of bubbles occur, resulting in the rupture of agglomerates. Time, power and irradiation modes (continuous or pulsed) are the key parameters affecting dispersion quality in an ultrasonic bath. Nguyen et al. [16] showed that there is an optimum power input in ultrasonication: past a certain point, a higher vibration amplitude will not improve dispersion quality, but will actually increase the re-agglomeration rate. High-speed revolution shearing, milling [14], and high-pressure homogenizers [13] are the main mechanical dispersion approaches. In milling, dispersed nanoparticles are introduced from the bottom of the mill in a slurry. Agglomerates are broken by passing through the stirrer, impinging the beads, and being stirred by rotating pins. In the upper part of the mill, the beads and slurry are separated by centrifugation and the dispersed-particle containing slurry is discharged. Similar to sonication, Inkyo et al. [14] also indicated that there is an optimum time for milling, after which re-agglomeration occurs.

While liquid phase techniques are generally effective at dispersing nanoparticles, they do not directly address the issue of re-agglomeration.

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Table 1
Comparison of different nanoparticle dispersion methods in liquid and gas phases.

Techniques	Mechanism of dispersion	Advantages	Disadvantages	References
Ultrasonication	Acoustic cavitation	Simplicity. Applicable for a wide range of nanoparticles. Efficient for high particle concentration. Wide range of liquid viscosities. Possibility to control more parameters (amplitude, irradiation mode, pressure, temperature, viscosity, and concentration)	Re-agglomeration problem. Contamination of nanoparticles, additives required (thermal stability issues). Downstream separation problem	[16] and [12]
Bead milling	Friction and shear flow	Applicable for a wide range of nanoparticles. Multi-pass and continuous. Scalable	Complex design. High energy input. High polydispersity. Damaging to particle structure. Additives required (thermal stability issues). Addition of impurities to nanoparticles	[14] and [15]
High-pressure homogenizer		Well established technology. Effective dispersion. Suitable for thermos-sensitive material Reproducible. Scalable.	Extremely high energy inputs. High polydispersity	[13] and [17]
Nozzle and orifice	Shear flow and collisions with other particle, clusters, and walls	Controllable. Low cost and energy. Continuous. Scalable	Not able to produce sub-100 nm particles. Re-agglomeration issues	[7], [8], and [9]
Low pressure impactors	Shear flow through the nozzle and impaction	Well established. Ability to produce monodisperse powders. Low cost and energy. Continuous	Complex design. Extremely low pressures. Re-agglomeration issues	[4] and [10]
Fluidized bed	Acceleration and impaction between the powder that causes attrition	Well established. Possibility of pre- and post-treatment in situ. Scalable	Low dispersion energy. Difficult to produce sub-100 nm particles. Re-agglomeration issues	[5] and [6]
Rapid Expansion of Supercritical Suspensions (RESS)	Shear flow as result of rapid expansion and impaction as the powder enters the capillary tube	Possibility of pre- and post-treatment (e.g. ultrasonication and encapsulation). Benign de-agglomeration. Suitable for non-spherical nanoparticles (e.g. carbon nanotubes)	Difficult to control. Extremely high pressures (supercritical fluids).	[3] and [11]

To stabilize the suspension, additives must be supplied to provided electrostatic, steric and electrosteric repulsion effects. Surfactants are commonly used for this purpose, resulting in electrostatic repulsion between surfactant-coated nanoparticles [18] and reducing particle agglomeration caused by attractive van der Waals forces [19]. However, surfactants face several limitations, not the least of which is their thermal instability: surfactants can desorb from nanomaterials at relatively low temperatures (65–70 °C) [20], cancelling out their repulsive properties.

Gas phase methods have several advantages over liquid-phase approaches, such as the absence of solvent waste, the simplification of downstream separation, the feasibility of continuous processing, and the versatility with respect to particle material and size and structure [21]. The de-agglomeration of nanoparticles down to their constituent primary particles in the gas phase can be achieved by applying an external force larger than the interparticle forces. There are several methods to de-agglomerate nanoparticle clusters in the gas phase ranging from high energy (e.g. rapid expansion of supercritical suspensions [3] or low pressure single stage impactors [4]) to low energy (e.g. fluidized bed [5]). However, so far, the low energy methods have not been able to produce sub-100 nm particle sizes, and the high energy methods have significant scale-up issues because of their operating conditions. Indeed, the rapid expansion of supercritical suspension systems requires high pressures (1.9–7.9 MPa). Nurkiewicz et al. [6] presented a nanoparticle aerosol generator consisting of a vibrating fluidized bed with a baffle, a vibrating Venturi disperser and a cyclone separator. Although they discussed the de-agglomeration of nanopowders and preventing re-agglomeration (strictly through dilution), the investigation was mostly focused on generating nanoparticle aerosols at constant particle concentration over time to perform inhalation studies, limiting the scale-up potential. In order to break agglomerates in a controllable and scalable manner, further investigation is necessary, namely on interparticle forces and attaining the required de-agglomeration energy in a fluidized bed configuration.

In this work, we break the large fractal-shaped agglomerates of silica nanoparticles to smaller clusters continuously through use of a jet impactor-assisted fluidized bed (JIAFB). The force required to destroy the agglomerates is controlled by the gas jet velocity in the impaction zone. Calculating the impaction velocity determines the kinetic energy of particles upon impaction, making it possible to measure the theoretical fragmentation degree of nanoparticles. As agglomeration is a reversible phenomenon, in order to produce stable particles, reducing interparticle attraction and preventing the nanoparticles from re-agglomeration are inevitable. Therefore, following agglomerate destruction, the JIAFB includes a surface functionalization post-treatment, based on photo-initiated chemical vapor deposition (PICVD) [22], to ensure particle stability and prevent re-agglomeration (not reported here).

2. Experimental

2.1. Materials

Silica nanopowder manufactured by TEKNA™, via thermal plasma synthesis, was used for all fluidization experiments. Primary particles have an average diameter of 20 nm. The powder's specific surface area is 200 m^2/g, with a solid density of 2200 kg/m^3, and bulk density of 35 kg/m^3. As nanopowders are a strong absorber of humidity, the particles were dried at 140 °C and −70 kPa vacuum before any fluidization experiments. Argon was used as fluidizing gas and air was used as jet and diluter gas in all experiments.

2.2. Experimental set-up

The JIAFB consists of a jet and an impaction plate that are placed inside the bed, as well as an air-driven venturi pump installed on the outlet of the column (Fig. 1). The JIAFB uses a high-speed jet to accelerate the fluidized agglomerates onto the impaction plate. The venturi pump provides a vacuum in order to entrain out broken particles, as well as minimize re-agglomeration of dispersed particles by diluting the outlet flow and send it to the particle sizer (dilution ratio of 1:25). The particle size and mass concentration produced by the JIAFB was measured utilizing an in-line scanning mobility particle spectrometer (SMPS), operated in aerosol mode. A calibrated rotameter was used to set the inlet flow rate to the particle sizer. A cylindrical quartz tube was used as the fluidized bed with an internal diameter of 8 mm and a height of 60 cm. After passing through a porous glass wool distributor,