Experimental Study on Fluidization Behavior of Geldart B and D Particle Systems in Deep Tapered Beds

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Abstract

For large Geldart B and Geldart D particles, the working height attainable in cylindrical bubbling fluidized beds is typically limited by growth of bubbles. Bubbles coalesce rapidly in these systems, and as a consequence, violent motion associated with bubble breakage at the freeboard can lead to uneven fluctuating forces on the vessel. For Geldart D particles, spouted beds are typically employed to avoid large bubbles, but in a reacting system, use of a spouted bed does not allow uniform solids contact with the reagent gases. While baffles can be used to reduce the size of bubbles in Geldart B systems, for the intended application, erosive wear could not be tolerated for the scale-up opportunity. Likewise, reductions in average particle size or superficial velocity or decreases in bed height to accommodate larger bed cross-section were all undesirable for the intended operating targets. For the latter, the available external surface area for heating from the wall is also compromised when compared with the heat duty needed for scale-up.

An exploratory investigation of bubbling fluidization with tapered geometries was conducted for Geldart D and large Geldart B silicon particles. Two separate, tapered fluidized bed systems, one 8-in.×15.5-in. (20.3 cm×39.4 cm) diameter and a second 17.5-in.×40-in. (44.5 cm×101.6 cm) diameter, were constructed from plexiglass and used for experimental studies of bubble growth, bubble size, bed density, and aggregate fluidization behavior of these particles. Acoustical monitoring of pressure fluctuations and accelerometer-based vibration measurements were employed in addition to optical bubble probes and high speed cameras to characterize the bed hydrodynamics over distinct conditions of gas superficial velocity and bed height. Transition conditions for slugging behavior were identified for two distinct particle size distributions. Modifications to overall fluidization behavior were demonstrated with design changes to gas delivery, thus allowing enhancement of the overall bed inventory while avoiding the slugging regime. Comparisons of the experimental data with calculations of fluidization behavior with commercially available simulation software are also presented.

Introduction

Reducing cost of high purity silicon for the production of photovoltaic energy via silicon-based solar cells has been a topic of interest since the late 1970s, but with the continued growth in this market sector for energy production, global interest in this material has grown greatly, drawing attention from industry and academia [1]. Fluidized bed reactors used for the production of silicon are more efficient on energy utilization than the most widely practiced Siemens-type bell jar decomposer process, thus creating a product form having ability for reduced energy payback for solar cells and lower carbon dioxide emissions for energy production [2, 3, 4]. However, scale-up of these systems as noted by Filtvedt et al. is an endeavor due to the operating regime of temperature and chemistry required for the process balanced against the need for maintaining a high product purity standard (typically targeting ppba levels of dopant and metal impurities) [2]. Mitigation of fines formation within the bed, management of wall deposition, delivery of energy for process heating, gas injection design, and contamination control with product handling all present unique challenges for process engineers.

A key barrier in the scale-up of the fluidized bed deposition process is the management of bubbles as the diameter of the reactor is increased. For the commercial application of high purity silicon (particle density = 2.33 g/cm³), it is desirous to have larger particles greater than 1 mm to reduce contamination potential, but this operating regime yields beds having either larger Group B or Group D
Geldart particle behavior. Slugging behavior is easily realized with these particle groups, and operation in this regime creates problems for scale-up. Bubbles tend to grow unbounded as emulsion phase gas transfers into the bubbles, leading to significant pressure fluctuations in the system and uneven forces on the vessel wall. To deal with this problem, beds containing Group D solids are typically either run shallow or run in a spouted regime, which sacrifices some gas-solid contact efficiency. Alternatively, the design basis can restrict the average particle size or maintain smaller bed cross-sections which affects net throughput. However, as the fluidized bed deposition reactor is scaled to improve economy of capital investment, there is also the need for more energy input. For instance, the maximum heat flux from the wall to the bed can be limited by the allowable thermal stress associated with the wall material of construction and heat delivery method. Internal heaters and heat exchangers have been proposed in the art to supplement energy input, but these add complexity not to mention challenges with maintaining product purity [2]. With limitations on heat flux, increases to the FBR diameter result in larger height, but bubble growth rate increases resulting from more bed height can lead to excessive slugging in the FBR. While mechanical solutions such as baffles can be used to break bubbles, in the polysilicon deposition application, implementation of a robust, erosion-resistant baffle design that likewise doesn’t induce contamination of product is difficult.

Tapered Geometry

An approach for consideration was use of a tapered geometry. In a tapered system, the gradual increase in bed cross-section allows for gas exchange between the rising bubbles into the surrounding emulsion phase, thereby limiting the growth of bubbles. Angles for the taper are kept small, typically less than 10 degrees, to allow a gradual transition in superficial gas velocity with height. Tapered beds were first used by the Union Carbide Nuclear Company for batch reduction reactions of small dense uranium ore particles, allowing reduction in slugging on the small 5-in. (12.7 cm) diameter reactor [5, 6]. This concept has been extended into other applications such as biological waste water treatment using three phase beds and coal gasification processes, but little work has been reported in the literature regarding their operation, with most studies focusing on prediction of minimum fluidization velocity [7, 8, 9]. Discussion of slugging behavior in these systems has been more limited. Maruyama and Koyanagi describe some experimentation on slugging behavior for small Geldart B particles in a tapered bed for various apex angles for the taper, but the inlet section of the bed is quite small (only 3 cm×3 cm), which limits its applicability to Group D systems under consideration [10]. To evaluate suitability for deep tapered beds at targeted throughput, a series of experimental investigations were conducted to evaluate hydrodynamic behavior, and comparisons were made with a CFD model.

Experimentation

Validation studies on the bed hydrodynamics were conducted using principles of fluidization hydrodynamic scaling as described by Horio et al. [11]. The scaled beds for the prescribed geometry were tested with model representatives under hydrodynamically equivalent conditions using air as the fluidizing gas to those operating conditions required in a silane or chlorosilane-based deposition process. Two separate, tapered fluidized bed systems, one 8-in.×15.5-in. (20.3 cm×39.4 cm) diameter and a second 17.5-in.×40-in. (44.5 cm×101.6 cm) diameter, were used for experimental studies of bubble growth, bubble size, bed density, and aggregate fluidization behavior of these particles. Both systems were constructed of plexiglass in most areas to allow for visualization of the bed behavior during the experimentation. Schematics of both systems are depicted in Figures 1 and 2 respectively.

During the experimentation, differential pressure measurement devices were used to help characterize bed behavior. Since the fluctuation behavior is dynamic with bubbles growing in size and breaking apart at the freeboard, the preferred approach for characterizing their magnitude is the time series-average of the standard deviation of pressure fluctuations. The standard deviation of pressure fluctuations is proportional to the excess gas velocity and hence a measure of aggregate bubble size for the largest bubbles formed in the system. As mentioned by Davies et al., this measurement can be used as
an inference tool for particle size monitoring [12]. In addition to differential pressure measurement, an accelerometer was also used to measure wall vibrations. Work in this area has been attributed to Cody et al. for detection of bed level, but this study focused on detection of slugging behavior [13].

A critical aspect for the acquisition of pressure fluctuation data is that the pressure measurement device be fast acting relative to the bed frequencies (which are usually below 10 Hz). Moreover, the conduits connecting the process to the pressure measurement device must be small enough that the vapor volume contained within the conduit does not act as dampening reservoir, which would result in false low measurements of the real pressure fluctuations. To alleviate this problem, conduit size is not recommended to be larger than 0.25-in. (6.4 mm) diameter nominal. For the experiments conducted in this study, the conduit size was held at 0.125-in. (3.18 mm) diameter and purges were maintained on the taps to reduce risk of plugging. The differential pressure over the lower 1 ft (0.3 m) of bed height was monitored and processed with a high frequency response data acquisition system; Validyne-series differential pressure transmitters were used in both cases, with the rate of response operating at 1 kHz for the 8-in.×15.5-in. bed and 200 Hz for the larger 17.5-in.×40-in. bed. Additional taps existed higher in the bed. An accelerometer (Model: Kistler K-Shear 8712A5M1) was also placed on the 17.5-in.×40-in. diameter vessel at a distance of 56 inches (142.2 cm) above the top of the gas distributor, and this device was coupled to a data acquisition system to acquire and process the acoustical signals. The standard deviation of the accelerometer signal was also calculated over a time averaged period of 60 seconds.

**Experimental Results and Discussion**

8-in.×15.5-in. Tapered Bed Study

A mixture of silicon particles was charged into the system shown in Figure 1. The average particle size distribution had a surface-to-volume (Sauter) mean diameter of 1515 μm, which made the system fall in a Geldart D regime. Solids inventory was varied between 55 lb (25 kg) and 129.5 lb (58.7 kg). Slugging was not observed, in the tapered system and the time-averaged standard deviation of differential pressure was below 5 inches H2O (1.25 kPa) across the measurement span. In contrast, experiments with an 8-in. cylindrical bed showed a clear transition to slugging behavior for pressure fluctuations in excess of 5 inches H2O. The geometric transition from the gas distributor to the tapered section was also varied. While it is possible to have a straight section just above the distributor, it is recommended that the length be kept small (L/D<0.5) since bubbles form quickly in Geldart D regime. It was observed that if a large bubble formed prior to entering the tapered section, the bubble would not dissipate and a central moving slug would remain.

17.5-in.×40-in. Tapered Bed Study

For the second system depicted in Figure 2, two separate particle size distributions were tested (Sauter mean diameters of 974 μm and 1263 μm respectively). In the first study, traditional slugging was not observed for the 974 μm particle size distribution at bed inventory of 1932 lb (876 kg) and the standard deviation of differential pressure went as high as 10 inches H2O (2.49 kPa) over the measurement span. When the bed inventory was increased further to 2919 lb (1324 kg) corresponding to a fluidized height approximately 8.5 ft, some violent bubble breaks were observed. For the 1263 μm mixture, slugging was clearly observed when the standard deviation of pressure fluctuations exceeded 7 inches H2O (1.74 kPa). For this larger distribution, slugs were observed at the bottom of the bed over most operating velocities of interest. A second set of gas injection nozzles is placed within the tapered section at an elevation of 62 inches (157.5 cm) above the top of the gas distribution plenum. Fluidization was greatly improved for this particle size distribution (see Figure 3) and improvements to the system behavior were observed as the percentage of total gas flow was increased (up to 40% of total flow). At the highest bed inventory, a small slug was noticed with secondary aeration in the bottom of the system, but this quickly dissipated so an effective threshold of 8 inches H2O (1.99 kPa) was deemed tolerable while operation without aeration was clearly intolerable (see Figure 4). Moreover, in a real system, there will be a thermal transition of the
Gas due to heating which should result in less excess gas velocity than what was physically simulated in the model system.

No change in bed density was observed as a function of aeration gas flow when the bed was in a fluidized state. Bubbles were also characterized using an intrusive bubble probe at the 6 foot (1.83 m) elevation position. Even though the measured pierced lengths (vertical dimension) of the gas bubbles are in the range of 8 to 20 inches (20.3-50.8 cm), they don't appear to bridge across to form gas slugs when the staged aeration is activated. Similarly, use of staged aeration provided benefit for the 974 μm Sauter mean population. This was observed in accelerometer vibration and reduced standard deviation of pressure fluctuations. These results suggest that for most operational flexibility, use of secondary gas injection is a strong benefit. Mechanical implementation of secondary gas injection does present some challenges for a system operating at temperature with chemistry. These nozzles should be shrouded and cooled to prevent silicon deposition build-up, and they should enter the reaction vessel horizontally between sections of induction coil or resistance heaters.

Experimentation with the accelerometer showed that for constant bed inventory and gas velocity, there were higher bed vibrations (measured acceleration) for the 1263 μm population over the 974 μm population. This behavior suggests the ability to infer particle size information from bed inventory and operational velocity feedback measures when coupled with accelerometer data.

**Modeling and Simulation**

Computational fluid dynamics studies of tapered beds have been reported in the literature. Sau and Biswal completed a study of Geldart D particles in a 1.97 in.×5.32 in. (5 cm×13.5 cm) diameter system where experimental results were compared with an Eulerian-Eulerian 2D-axisymmetric model featuring Gidaspow’s fluid-solid drag law with computations performed with ANSYS/Fluent® software platform [14]. Bed pressure drop and bed expansion were compared with the model yielding slightly lower predictions of bed pressure drop but poorer agreement on bed expansion. Duangkhramchan *et al.* at Ghent University showed improved agreement with an Eulerian-Eulerian flow model with multiple solid phases to better approximate the distribution of solids in a 5.5 in.×11.8 in. (14 cm×30 cm) tapered bed fluidized from air [15]. Their application focused on Group B glass bead powders using a half round, 3D model using Fluent® software as the computational platform. The modified Gidaspow drag law model was determined by the researchers to give the best model representation of solids volume fraction for their experiments. However, the model looked at experiments with very shallow bed heights compared with the inventory capability of the system. Liyan *et al.* repeated simulations of the Ghent University experiments using a two fluid model featuring a different drag law, with this one being based upon a tapered bed equivalent Ergun equation, coupled with second order moment frictional stresses [16]. However, this method results in a drag law dependent on tapered angle of the walls instead of a geometric independent drag law.

For this computational study, the multiphase particle in a cell method (MP-PIC), which is an Eulerian (fluid)-Lagrangian (solid)-based model, was applied to a three dimensional model of the 17.5-in.×40-in. tapered bed [17, 18]. In the simulations, the Wen and Yu and Gidaspow fluid-solid drag laws were both tested. Computations associated with these models were completed using CPFD Software’s Barracuda® software. Use of aeration gas at the locations was modeled explicitly for the 17.5-in.×40-in. diameter system. Of particular interest, the time-averaged standard deviation of differential pressure was compared between experimental and simulation conditions for several superficial gas velocities using the 1263 μm population (see Figure 5). Neither drag law appeared to match bubble behavior adequately across the length of the bed although the Wen and Yu drag law had better agreement near the top of the bed.
Summary

Use of staged gas injection in the scale-up of tapered beds improved overall operability for large Geldart D particles, allowing for additional bed height. The use of accelerometer measurements provided good agreement with pressure fluctuation measurement data and it showed clear differentiation between the average particle size for fixed bed inventory and gas throughput. CFD models of this system have need for further development to properly predict bubble growth dynamics and pressure fluctuations.

References

Figure 1: Schematic for 8-in. × 15.5-in. Diameter Plexiglass Tapered Bed Apparatus Indicating Pressure Measurement and Test Probe Locations Accompanied by Photograph of Test Unit at Particulate Solids Research, Inc. (Chicago, IL USA)
Figure 2: Schematic for 17.5-in.×40-in. Diameter Plexiglass Tapered Bed Apparatus Indicating Pressure Measurement and Test Probe Locations Accompanied by Photograph of Test Unit at Particulate Solids Research, Inc. (Chicago, IL USA)
Figure 3: Effect of Staged Aeration upon Standard Deviation of Pressure Fluctuations (1263 µm Silicon Beads in 17.5-in. x 40-in. Tapered Unit with 1932 lb bed inventory)

Figure 4: Effect of Bed Inventory upon Standard Deviation of Pressure Fluctuations (1263 µm Silicon Beads in the 17.5-in. x 40-in. Tapered Unit with and without Staged Aeration)

Figure 5: Comparison of Standard Deviation of Differential Pressure Fluctuations (Experimental Measurement vs. CFD Simulation of 17.5-in. x 40-in. Tapered Bed Study of 1263 µm Silicon Beads as a Function of Aeration Flow; Fixed Bed Inventory = 1932 lb)