PROCESSING COHESIVE FINE PARTICLES IN A SPOUTED BED VIA MIXING WITH SPOUTABLE MEDIA

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Abstract – A small cold flow spouted bed was constructed and utilized for the study of spouting cohesive Geldart class C powders in the presence of larger spoutable media. Tests were conducted with various mixtures of 871 µm HPDE pellets and 0.7 µm hematite. Test results suggest that the addition of the cohesive hematite to a bed of larger HDPE resulted a more evenly dispersed average pressure drop profile as a function of gas velocity. In addition, it was observed that the addition of hematite lead to a decrease in the percentage of gas-supported solids mass within the bed, to a minimum of approximately 65% supported mass at mixtures containing between 30-50% hematite by weight.

INTRODUCTION

Many industrial processes involve the conversion or reduction of solid materials via non-homogeneous reactions between the solid material and a surrounding gaseous or liquid medium. In the case of solid-gas reactions, fluidized beds are perhaps one of the most popular reactors because individual particles are suspended within the gaseous phase, which provides excellent surface contact for the desired reactions to take place. However, not all types of solid particles can be easily fluidized. For example, very fine particles (Geldart class C particles) are more susceptible to inter-particle cohesion forces, such as Van der Waals, capillary, and electrostatic forces, and tend not to fluidize. Instead, dense beds of these cohesive particles tend to agglomerate and form cracks or channels that allows the gas phase to bypass the solids with very little contact between the two (Fan and Zhu, 1998).

This behavior has in the past imposed limitations upon the minimum particle size for gas-solid reactions utilizing fluidized bed reactors. These size limitations tend to have a negative impact upon the reaction kinetics for the desired chemical processes occurring within a given reactor vessel. In general, apparent kinetic rates increase with decreasing particle size due to smaller particles having larger surface area-to-volume ratios. Because of this, smaller particles are preferred from a kinetics standpoint, but below a certain size threshold, particles transition from Geldart class A to the cohesive Geldart class C, and thus suffer from the difficulties discussed in the previous paragraph (Geldart et al., 1984). This then forces a tradeoff between ease of fluidization and effective reaction rates within the gas-solid reaction system.

Consequently, the most common technology currently in use for reactions involving Geldart C particles utilize either rotating drums (i.e. rotating kilns) or mechanical agitators to mix the process reactants. However, these methods are not without their own limitations. These mechanical methods involve the use of moving parts often operating at high temperatures, where the likelihood of mechanical failure is increased; thus, requiring potentially expensive maintenance and operating costs. Additionally, since these mechanical methods usually entail a dense bed of solids that are in direct contact with surrounding particles, there is less contact area between the solid and gas phases, which can lead to a rate-limiting condition that reduces the extent of chemical conversion.

Alternatively, there has been some research conducted to explore the idea of utilizing vibrated fluidized beds for fluidization of cohesive particles. Morooka et al. (1998) studied fluidity of submicron-sized powers of Ni, Si_3N_4, SiC, Al_2O_3 and TiO_2, as well as agglomeration. They found fluidization of these possible at high gas flow rates after the bed was vibrated enough to break up particle agglomerations, but $CaCO_3$ and ZrO_2 were not fluidizable under these conditions and quickly re-agglomerated and led to bed defluidization.

Mawatari et al. (2002) conducted a study of particle diameter on fluidization under vibration using glass beads ranging from 6 (Geldart group C) to 100 (Geldart group A) microns. Through varying the intensity of vibrations within the bed, they found that the velocity for minimum fluidization of the cohesive particles decreased with increasing vibration intensity. They also found that smaller particles led to complex flow patterns within the bed due to the formation of agglomerations of multiple sizes that made it difficult for vibrations to propagate into the bed.

In a follow-on work, Mawatari et al. (2005) studied the upper gas velocity limit for vibro-fluidization of 6μm glass beads. These upper and lower gas velocity limits were defined as the gas velocity above which channels formed within the bed. The authors found that this maximum gas velocity varied depending upon whether tests were conducted with increasing or decreasing gas velocities, where the case of increasing gas velocities lead to higher maximum velocities before the re-emergence of channeling and defluidization of the particles.

Xu and Zhu (2005) studied the effects of vibration on the formation of agglomerations in beds of fine Talc and CaCO3 particles. They found that mechanical vibration cannot only facilitate breaking of agglomerates, but can also favor the growth of agglomerates if the vibrational intensity exceeds a critical level.

In each of the above studies, it was shown that while mechanical vibration of the fluidized bed can disrupt the formation of agglomerates and channeling in cohesive powders, thus leading to enhanced fluidization under some vibratory conditions, other vibratory conditions can lead to enhanced formation of agglomerates and channeling, ultimately leading to defluidization of the bed. As a case in point, the authors suspect that this is the reason that Marooka et al (1998) experienced re-agglomeration and bed defluidization while trying to fluidize $CaCO_3$ and ZrO_2.

Other researches have taken an alternative approach towards fluidizing cohesive powders. In these studies, the researchers attempt to tackle the problems resulting from cohesive particle agglomeration and channeling by introducing larger particles into the fluidized bed.

Alavi and Caussat (2005) studied the fluidization of Y_2O_3 powders, and the effects mechanical vibrators and stirrers, as well as addition of larger particles, on their fluidization characteristics. they found that the mechanical aids had little effect on the cracking and channeling prevalent during fluidization of these cohesive particles, but the addition of larger particles lead to acceptable fluidization conditions.

Bush and Workman (1981) submitted a patent application for a process incorporating fluidized and spouted beds for particle coating applications used to produce tantalum-coated alumina powders to be used for formation of capacitor anodes for electrolytic capacitors. In the proposed process, 3-micron diameter alumina particles were mixed with 120-micron alumina particles in mixture ratios ranging from 2-to-1 to 4-to-1. It was reported that the addition of these larger alumina particles reduced the agglomeration of, and channeling in, the smaller 3 micron powders sufficiently to allow for fluidization.

Similarly, Brooks (1985) submitted a patent application to produce a fluidization aid consisting of a tendrillar carbonaceous material. This material, a fibrous particle consisting of carbon fibers with a ferrous metal component, would be mixed with cohesive Geldart C powders to allow for fluidization, as well as with Geldart A and B materials as a fluidization enhancer.

An effort is currently under way at the National Energy Technology Laboratory (NETL) to develop an easily scalable chemical reactor system for processing of fine cohesive particles utilizing the mixing of a larger non-reacting fluidization aid material with the desired fine cohesive power reagent within a modular, quasi-2D rectangular spouted bed reactor. For this work, a study was conducted to examine the effects of different ratios of cohesive powder–to–non-reacting fluidization aid upon the minimum spouting velocity, bed pressure drop, and elutriation rate of fine material.

EXPERIMENTAL SETUP

The chemical reactor under development at NETL is based upon utilizing a spouted bed with a spoutable media to more easily fluidize the Geldart class C fine particles to improve mixing and contact area between the fluidizing gas and fine particles. Fig. 1 shows a conceptual sketch of a typical spouted bed. In a spouted bed, the fluidizing gas is injected into a dense bed of particulate material located at the bottom of the bed, which is typically conical in shape. The fluidizing gas forms a jet that creates a core of upwards moving gas (spout) within the bed of particles that pushes the particles located within the core up through the densely-packed particles, until they are ejected from the bed of particles into the freeboard region above the dense bed. Assuming the gas velocity in the freeboard region is less than the terminal velocity of the particles, the particles then fall back down into the dense bed region. In addition, as particles are carried upwards throughout the core region formed by the gas jet, additional particles are entrained into the bottom of the core from the surrounding area, commonly referred to as the annulus. This produces a circulatory motion with the particles located in the annulus region, as depicted by the arrows in Fig. 1.

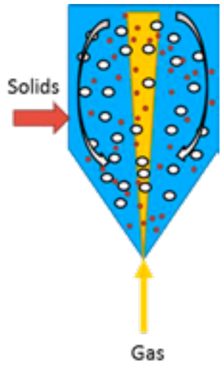

Figure 1: Conceptual diagram of a spouted bed

As previously stated, Geldart class C particles are typically considered to be unfluidizable due to the dominance of inter-particle cohesive forces, which leads to the fluidizing gas forming channels through the material, leaving the bulk of the solids unfluidized. Because of this, it is necessary to make use of a second solid particulate material that is more readily fluidized (or spouted in our case). When this secondary particulate material (hereafter referred to as the 'spoutable media') is introduced into a bed of Geldart class C particles, the media interacts with the gas flow and begins to exhibit the spouted bed behavior detailed in the previous paragraph. As the spoutable media is ejected out of the core and falls back into the annulus region, it collides with clumps (or clusters) of the more cohesive particles and breaks up these larger clusters. As the clusters of smaller, cohesive particles are broken up, the cohesive forces are overcome by other forces acting upon the particles, and they too eventually begin to exhibit spouting behavior within the bed.

A small-scale cold flow model of the spouted bed chemical reactor unit is shown in Fig. 2. Fig. 2(a) provides a process flow diagram, and the actual experimental unit is depicted in figure 2(b). the experimental unit has a cross section of 4 inches wide by 1 inch in depth, with a total height of approximately 32 inches. The lower section of the bed is sloped with an angle of 60°. Solids are loaded into the unit via a port located at the top of the bed, as well as through the side via a small plunger-like feed device (not shown). The spouting gas (air) enters the bed via a 3/8-inch diameter nozzle located at the bottom of the cone section, and exits through a pair of outlet ports located at the top of the unit. The rate of airflow into the unit is controlled via an alicat mass flow controller with a range of 0-1500 slpm. The gas exits the unit via exhaust ports located at the top of the unit, where any entrained solids are separated via filters. For collection of differential pressure data, pressure taps are located at multiple locations along the height of the unit. These are connected to a series of Setra model 239 differential pressure transducers with ranges of 0-30 in-H_2O. The signals from these transducers are sampled and recorded via Labview at a sample rate of 100hz.

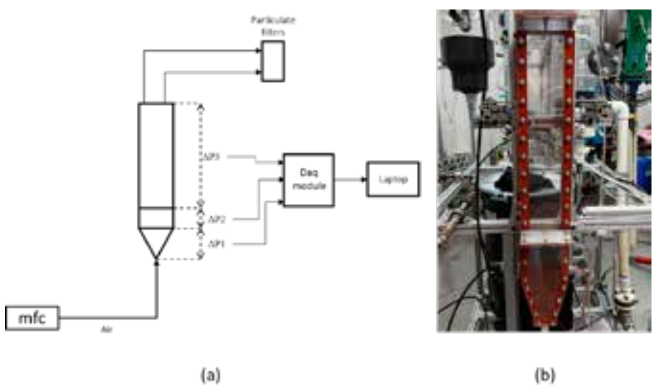

Figure 2:(a) Process and instrumentation diagram (b) cold flow spouted bed unit.

Experiments were carried out using a variety of static bed heights and mixtures of 0.7µm hematite and 871µm HDPE pellets. Table 1 provides the material properties of the hematite and HDPE.

Table 1: Material Properties

	HDPE	Hematite
Particle Diameter (µm)	871	0.7
Density (kg/m^3)	860	5300
Min. Fluidization Velocity (m/s)	0.17	1.96 x 10^{-10}

RESULTS AND DISCUSSION

Prior to conducting any experiments involving mixtures of the HDPE and hematite materials, a set of preliminary tests were carried out to establish baseline data for beds consisting entirely of either HDPE or hematite, as well as to explore what effect, if any, varying the initial packed bed height had on the minimum spouting velocity of the HDPE. Fig. 3 present the data obtained for the HDPE for initial packed bed heights of 4 and 6 inches, as well as for a 4-inch initial packed bed containing hematite. For the 4- and 6-inch packed bed HDPE cases, a typical average pressure profile for a spouting bed is observed. The initial linear increase in pressure drop corresponds to the formation of a dome of gas forming at the bottom of the bed. As this dome of gas expands towards the top of the packed bed, the pressure increases until it reaches a maximum value, increasing the gas velocity beyond this point leads to the formation of the spout that is iconic of a spouted bed flow regime. The point at which spouting occurs is denoted by the sharp decrease in the bed pressure drop. From the data presented in Fig. 3, the experimentally obtained minimum spouting velocities for the 4- and 6-inch packed beds of HDPE are 1.3 and 1.9m/s, respectively. While spouting was not observed in the case of the 0.7 µm hematite, the average pressure profile for a 4-inch bed of hematite is included for comparison.

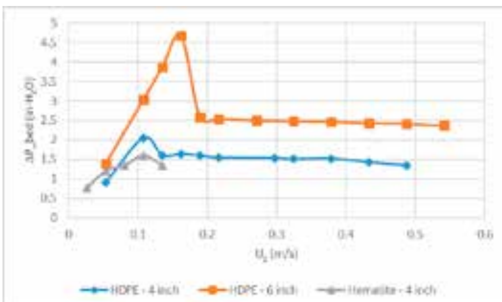

Figure 3: Average Pressure Profiles for various bed heights of unmixed HDPE and Hematite

Experiments were also conducted using mixtures of HDPE and hematite. For these experiments, HDPE and hematite were premixed in the following HDPE/Hematite mass ratios and loaded batch-wise into the spouted bed unit: 90/10, 85/15, 70/30, 60/40, 50/50. In each case discussed below, the spouted bed unit was loaded using a 4-inch bed height prior to introducing air into the system. Fig. 4 compares the average pressure drop profiles for the pure HDPE to the 90/20 and 85/15 HDPE/Hematite mixtures. As seen in figure, the addition of small amounts of the cohesive hematite has minimal effect on the average pressure drop at higher gas velocities. At lower gas velocities (prior to the minimum spouting velocity for the bed consisting solely of HDPE), the addition of hematite results in a decrease in the peak pressure, as well as decreases the slope of the curve prior to the peak pressure. Increasing the amount of hematite present in the bed, as shown in Fig. 5, leads to average pressure drop profiles that are nearly constant, regardless of gas velocity. This suggests that, in mixtures containing large amounts of the cohesive hematite, the gradual increase in pressure associated with the formation and growth of an internal dome of gas that expands towards the top of the packed bed as gas velocity increases is not present.

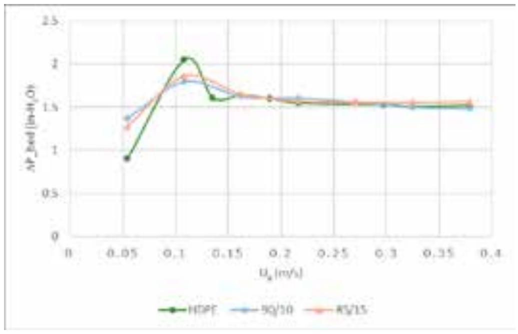

Figure 4: Average pressure profiles for 4-inch beds of pure HDPE and HDPE/Hematite mixtures of 90/10 and 85/15.

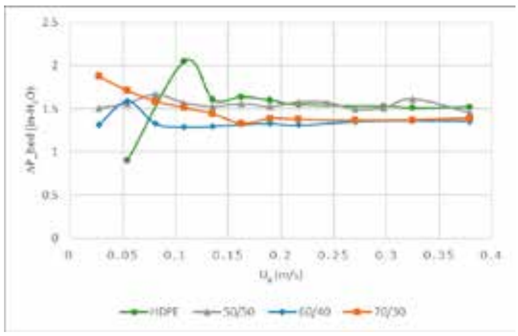

Figure 5: Average pressure profiles for 4-inch beds of pure HDPE and HDPE/Hematite mixtures of 70/30, 60/40 and 50/50.

Fig. 6 normalizes the average bed pressure drop information provided in Fig. 4 and Fig. 5 by dividing the average pressure drop by a pressure obtained from dividing the total weight of material in the bed by the cross-sectional area of the bed. Fig. 6 shows that, in the case of the 4-inch static bed of pure HDPE, the peak normalized pressure exceeds a value of 1.1 prior to the onset of spouting, beyond which approximately 85% of the solids within the bed are being supported by the fluidizing gas. As the cohesive hematite particles are added in ever larger ratios, the percentage of gas-supported solids decreases until appearing to converge upon a supported mass ratio of approximately 62%. This is more readily demonstrated in Figure 7, which shows the average of the normalized pressure drop for all velocities above the minimum fluidization velocity of the HDPE as a function of the mass percentage of cohesive fines (Fe_2O_3) in the bed. This is consistent with visual observation of the unit while in operation. While conducting experiments with the HDPE/hematite mixtures, it was observed that there is a region near each wall in which exists masses of solids which do not appear to interact with the gas flow, and furthermore do not appear to exhibit any recirculation of solids. This can be seen in Fig. 8. The geometry of the spouted bed unit used for the current series of experiments utilized a 60-degree cone section; however, additional tests will be conducted using other geometries to determine the effects of cone angle on this observed phenomena.

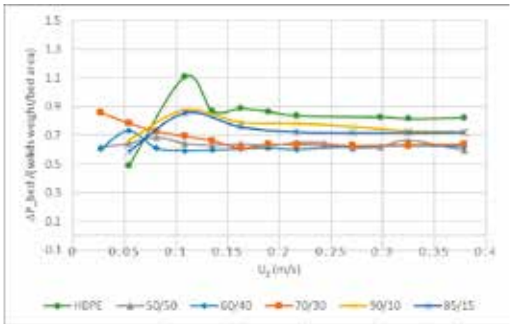

Figure 6: Normalized average pressure profiles for 4-inch beds of pure HDPE and HDPE/Hematite mixtures of 90/10, 85/15, 70/30, 60/40 and 50/50.

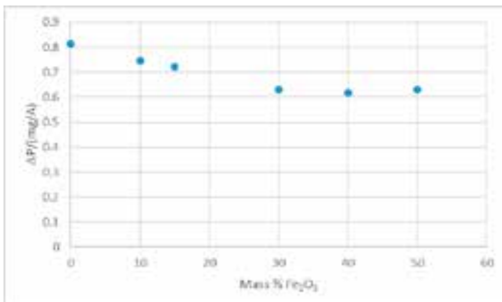

Figure 7: Average of normalized pressure values for all gas velocities beyond $U_g > U_{mf,\ HDPE}$ versus mass percentage of cohesive Fe2O3 powder in bed.

Figure 8: Image showing regions of apparent non-interactive particles (darker shaded areas) for the 50/50 HDPE/hematite mixture case. (u_g = 3.25 m/s)

Finally, Fig. 9 shows a comparison of the elutriation rate of the hematite vs. superficial gas velocity, U_g, for the 70/30, 60/40, and 50/50 mixture cases. The elutriation rates presented were obtained by differential weight measurements of the particulate filters downstream of the system gas exhaust ports, divided by the time length of the experimental run. In each case, no elutriation was observed prior to U_g = 0.16 m/s, which is approximately the measured minimum spouting velocity of the HDPE. Beyond that point, the elutriation rates are approximately linear with respect to gas velocity, and vary by the mixture ratio.

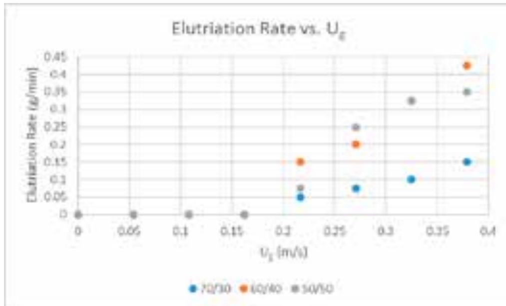

Figure 9: elutriation rate of hematite vs. superficial gas velocity, U_g, for the 70/30, 60/40, and 50/50 HDPE/hematite mixtures.

CONCLUSIONS

A small-scale cold-flow spouted bed device was developed for the study of the feasibility of spouting fine, cohesive Geldart class C powders with the assistant of a larger spoutable media as a precursor to development of a high temperature chemical reactor system for cohesive powders. Experiments were conducted in which different mixtures of 871 µm HDPE pellets and 0.7 µm hemitate powder were spouted over a range of superficial gas velocities (up to approximately twice the minimum fluidization velocity of the HDPE).

While the experiments showed that it was possible to obtain spouting of the cohesive hematite powder via mixing with the larger spoutable media (aka the HDPE), the resulting average pressure drop profiles for the mixtures containing larger percentages of the cohesive hematite lacked the traditional linear increase to a peak pressure, followed by a rapid decrease, followed by a level, horizontal pressure drop profile normally associated with spouting. Instead, the addition of the cohesive hematite smoothed out the pressure profiles, making them nearly linear and relatively unaffected by gas velocity, this becomes more pronounced for higher cohesive fines loading cases. Additionally, increasing the amount of hematite in the mixture lead to decreases in the percentage of solids supported by the gas flow due to formation of dead zones near the walls of the spouted bed reactor vessel. Based upon the normalized pressure drop data presented in fig. 6, it was concluded that approximately 35% of the solids within the spouted bed reactor is contained within these dead zones. To make a more efficient reactor system, additional testing will need to be conducted to explore, among other options, increasing the cone angle at the bottom of the spouted bed, as well as exploring the possibility of injecting additional air normal to the sloped reactor walls to help break up these dead zones.

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DISCLAIMER

The authors declare no competing financial interest.

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