Small-Scale Particle Interactions Are Having Significant Effects on Global Fluidized Bed Behavior†

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Abstract

Fluidized bed design and scale-up depends strongly on particle characteristics such as size, shape, and for Geldart Group A particles, the level of fines (particles smaller than 44 microns). However, recent research has shown that particle clustering has a significant effect on fluidized bed hydrodynamics which impacts how these units should be designed and scaled up. This is especially true with the estimation of the solids entrainment rate and the cyclone collection efficiency. The amount of fines, particle shape and surface morphology play a role on the level of particle clustering in a fluidized bed. The fine particles are an excellent conduit for moving charge as electrons or ions which appear to be the dominant mechanism of electrostatics for Geldart Group A material in a bubbling fluidized bed. This electrostatic force trades off with particle momentum relaxation and rotational to translation momentum transfer with regard to forming a particle cluster. The issue is the quantification of this effect so more precise calculations can be made with particle entrainment rates and cyclone collection efficiency. Preliminary work on particle shear in a packed and fluidized beds, suggest that particle clustering can be measured and may provide a quantifiable metric for the level of particle clustering.

Keywords: powder, particle, fluidization, entrainment, clustering, cohesive forces

1. Introduction

In the design and operation of fluidized beds, particles are assumed to behave independently of each other which results in a fluid-like state once all the particles have been suspended by the fluid. In other words, the factors responsible for dense granular flow such as particle shape, surface roughness, and stress chains nearly disappear once the particles are fluidized. It’s a common convention that has limited our understanding of fluidized bed hydrodynamics.

Through the years, this common convention has been challenged. Particle clustering has been postulated as early as in the 1890’s where fine-grained granular materials were observed to break up into “droplets” much like a liquid stream (Khamontoff, 1890). Wilhelm and Kwauk (1948) proposed that Geldart Group A particles clustered together, which resulted in the smooth fluidization observed just after the minimum fluidization velocity was achieved. This homogeneous or smooth fluidization is only observed for Geldart Group A particles below the minimum bubbling velocity. Larger particles, such as Geldart Group B particles, exhibit bubbling fluidization at the onset of fluidization. The smaller Geldart Group A particles were viewed as being responsible for particle clustering, which is not evident with larger particles. Geldart and Wong (1987) expanded on this by adding even smaller Geldart Group C particles to a fluidized bed of Geldart Group A particles. The result was a marked decrease in the measured entrainment rate. They suspected that the smaller particles or fines were adhering to the larger particle.

A study by Baeyens et al. (1992) was in agreement with Geldart and Wong’s work. Baeyens et al. noted that particles had to be smaller than 40 microns to exhibit this effect on entrainment. This maximum particle size was called the critical particle size and denotes the point where interparticle forces exceed hydrodynamic forces. Subbarao (2010) provided an expression for determining the cluster size which was inline with the findings of Baeyens et al. Jayaweera et al. (1964) found these clusters were stable over a range of two to six particles in size. Clusters larger than six particles were found to split and...
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1. Introduction

Particle clustering is an important phenomenon in fluidized beds, affecting the behavior of the particles and the overall performance of the fluidized bed. Fortes et al. (1987) observed that similar-sized clusters which were stable up to a Reynolds number of 1800.

Cocco et al. (2010) used a high-speed camera connected to a customized boroscope to reveal these particle clusters in the freeboard region of a fluidized bed containing fluidized catalytic cracking ( FCC ) equilibrium catalyst with a median particle size of 72 microns and 12 % fines (defined as particle with a size smaller than 44 microns). In fact, 30 % of the particle behavior in the freeboard was found to resemble particle cluster formation with an average particle cluster size of 11 particles. These clusters tended to be small particles or fines adhering to one or two larger particles. This finding was also consistent with similar observations in a fluidized bed of polyethylene powder where 75 % of the particle flow in the freeboard was in the form of particle clusters.

However, Kaye and Boardman (1962) noted that the concentration of particles in a freeboard tends to be too low for the formation of a significant amount of particle clusters. Solids concentrations needed to exceed 0.05 % which is unlikely in a freeboard of a bubbling fluidized bed. Matsen (1982) also concurred that cluster formation and the size of the cluster formation are strongly dependent on the solids concentration.

The same study by Cocco et al. (2010) used the high-speed camera with a boroscope to reveal that particle clusters existed in the fluidized bed as well as above the fluidized bed. The region surrounding a passing bubble showed that most of the particles were behaving as weakly-bound clusters. Cocco et al. suggested that perhaps clusters are formed in the bed and, in part, are subsequently entrained into the freeboard. Particle concentrations in the fluidized bed certainly exceed the criteria of Kaye and Boardman.

Hays et al. (2008) observed that entrainment rates were dependent on fluidized bed height. The taller the bed, the lower the entrainment rate. It was suggested that the reduction in entrainment rates with increasing bed heights was due to a longer growth time with the deeper bed for particle clustering. Hays et al. also added baffles into the fluidized bed and noticed that the entrainment rate increased in a system where small particles (less than 44 microns) were present. The baffles were believed to be responsible for breaking up these weakly-bound clusters. For beds with larger particles, there were not enough particle clusters to exhibit this effect. Both studies further support that particle clusters are mostly formed in the fluidized bed, and that the resulting entrainment rates and particle clustering in the freeboard are the result of particle clusters in the bed becoming ejected into the freeboard.

However, without understanding the underlying nature of the formation of particle clustering, it would be difficult to capture the hydrodynamics of a fluidized bed. This is more important now than in the past as more fluidized bed challenges are being addressed with computational fluid dynamic (CFD) models. In the past, fluidized bed design and operation was accomplished using empirical correlations which inherently contained the role of particle clustering in the experimental data. With CFD, the model is more fundamentally based, but it is based on the particles as being independent entities which is an incorrect assumption for many systems.

2. Impact of particle clustering on fluidized bed design

The lack of understanding of particle clustering in fluidized beds has complicated the scale-up of these unit operations. One of the key operating costs with fluidized beds are the solids loss rates. In catalytic systems, catalyst loss rates can amount to millions of dollars per year if a fluidized bed is not optimized. Fluidized bed design needs to consider particle attrition and particle collection to manage the loss of solids, which requires knowing the entrainment rate in the freeboard and the grade efficiency of the cyclones.

The entrainment rate is needed for cyclone and dipleg sizing. Under-prediction of the entrainment rates could lead to undersized cyclones prone to dipleg flooding. Over-prediction of the entrainment rate leads to an over sized-dipleg which can result in plugging because of too low of a solids flux moving down the dipleg.

Yet, entrainment rate predictions can be orders of magnitudes different, especially for Geldart Group A particles. Many empirical correlations, which were developed with experimental data already capture the clustering effect, inherently. However, such correlations are system specific. This certainly seems to be the case with the disparity of the entrainment rate from published correlations (Chew et al., 2015). Fig. 1 illustrates this discrepancy for the entrainment rate prediction using six entrainment rate correlations for FCC powder with 9 % fines in a fluidized bed at a superficial gas velocity of 1 m/s. For the small particles, entrainment fluxes were more than two orders of magnitude different, as shown in Fig. 1. Even the trends differ with some correlations showing entrainment significantly increasing with decreasing particle size while other correlations level off. It is likely that the lack of consideration or enough consideration for particle clustering could be a key issue with the discrepancies in these entrainment rate correlations (Chew et al., 2015). Furthermore, such differences can also be found in the calculations of the transport disengagement heights (Cahyadi A. et al., 2015).

Particle clustering can also lead to higher than expected cyclone collection efficiencies. It is hard to see the down
side of this scenario, but consider what happens when new cyclones are added to this fluidized bed with higher inlet velocities. Higher collection efficiency would be expected but that is not always the case. As shown in Fig. 2 for a 0.43-m diameter cyclone used in the collection of titania particles, higher inlet velocities resulted in a lower collection efficiency, not higher as expected. The higher inlet velocities were postulated as breaking up particle clusters with the additional trauma to the clusters. These clusters were preserved in the cyclone at the lower velocities. As a result, cyclone collection was influenced by smaller particles at the higher velocity. At the lower inlet velocities, a particle cluster was retained resulting in the cyclone “seeing” a larger “particle” and providing a higher collection efficiency.

Until there is a better understanding of the mechanism of cluster formation and how these clusters behave in fluidized operations, such discrepancies as those shown in Figs. 2 and 3 will continue. Fig. 3 illustrates this concept for the simulation of a 0.9-meter diameter fluidized bed of FCC particles with 9% fines. As shown in Fig. 3, Barracuda CFD simulations resulted in 20X higher entrainment fluxes than those observed experimentally for a 0.9-meter diameter fluidized bed of FCC catalyst with a 1.2-meter static bed height. Most commercial CFD codes do not account for clustering unless invoked with a drag correction (Yang et al., 2004). Hence, the entrainment rates can be significantly over predicted when CFD is used. Reasonable changes in the drag model may not help either. Fig. 3 illustrates this using a Wen and Yu drag model with a 0.8 or 1.0 multiplier. The entrainment rates from these simulations were relatively unchanged with respect to the differences with the experimental data. The experimental data are based on the experimental procedures outlined in Issangya et al. (Issangya et al., 2013).

3. Impact of fines levels on particle clustering

As noted by Baeyens et al. (1992), particle clustering in
A fluidized bed was the result of particles with a size smaller than 40 microns. Hays et al. (2008) observed this with a 0.16-m diameter fluidized bed of FCC catalyst (Geldart Group A, $d_{50} = 72$ microns, $\rho_p = 1500$ kg/m$^3$). As shown in Fig. 4 where grating-like baffles were added in the bed at 1.6 and 2.5 feet (0.5 and 0.75 meters) above the distributor plate, the entrainment rate was found to increase when superficial gas velocities exceeded 1 m/s. Hays et al. proposed that the higher velocities provided enough shear to break up particle clusters that existed in the bed. Yet, when larger coke particles (Geldart Group B, $d_{50} = 150$ microns, $\rho_p = 15600$ kg/m$^3$) were used in this unit, little differences in entrainment rates were observed over the whole range of superficial gas velocities, as shown in Fig. 5.

Direct evidence of particle clusters was observed and reported by Cocco et al. (2010). Using a modified borescope connected to a high-speed Phantom V7.2 camera, Cocco et al. were able to resolve particle clusters well above the bed in the freeboard. The borescope could capture particle and cluster flows beyond the influence of the wall where electrostatics could be more significant. Experiments were performed in a 0.15-m diameter fluidized bed at a superficial gas velocity of 0.6 m/s. As shown in Fig. 6, most of the clusters consisted of small particles or fines attached to larger particles. Clusters of only large particles were not typically observed.

Yet, Kaye and Boardman (1962) proposed that particle clustering becomes significant in many systems only where solids concentrations exceed 0.05 %. Such particle concentrations are higher than typically measured in the freeboard region of bubbling fluidized beds at superficial gas velocities of 0.6 m/s. The particle clusters may not be forming in the freeboard to any significant degree, but in the fluidized bed itself. This was confirmed in Cocco et al. (2010) with high-speed video measurements in the fluidized bed. As shown in Fig. 7, particle clustering is prolific in a fluidized bed of FCC catalyst particles.

Fig. 4 shows a selected frame from high-speed video captured in a fluidized bed of FCC particles using a modified borescope. The image in Fig. 7, is after a bubble passed by the probe where solids volume fractions concentrations are less than that of the bed emulsion. Nearly all the particles were behaving as clusters and small particles appear to have an integral role in the cluster formation. This level of particle clustering has a significant impact on the entrainment rate which is a product of the bubble bursting on top of the bed (Wen & Hashinger 1960).

It is unknown whether the clusters exist in the denser emulsion phase and get ejected into the less dense bubble phase, or if the clusters are solely a product of the bubble phase in the fluidized bed (Cocco et al., 2010). In the first
case, clustering is prolific and bed hydrodynamics may be strongly coupled to the level of particle clustering. In the second case, clustering may only affect entrainment rates but not bed hydrodynamics.

Hays et al. (2008) also reported that the entrainment rate was strongly dependent on the fluidized bed height. Fig. 8 shows the measured entrainment rate from a 0.15-m diameter fluidized bed for FCC catalyst powder with 5% fines at varying bed heights (Cocco et al., 2010; Hays et al., 2008). With the cyclone located well above the transport disengagement height for all cases, the entrainment rate was found to decrease significantly. A doubling of the bed height resulted in a 3.5 times reduction in the entrainment rates. Taller fluidized beds of FCC catalyst particles resulted in lower entrainment rates than shorter beds. Cocco et al. (2010) suggested that a taller bed provides a longer residence time in the bed for the clusters to get larger. What is unknown is how the concentration of fines in a fluidized bed changes the slope of this relationship.

Fig. 7 illustrates direct evidence of particle clustering in a fluidized bed. Figs. 4 and 5 show that particle fines (particles smaller than 44 microns) promote the formation of particle clusters which is not unexpected. Fines mobility can be significant in a fluidized bed and serves as a conduit for promotion of particle clustering. However, how do fines promote this particle clustering?

4. Clusters and cohesive forces

The questions to address are how do these clusters affect key design parameters such as the entrainment rate and cyclone efficiency. Specifically, what are the underlying forces that promote clustering in a fluidized bed? Possible mechanics for the formation of particle clusters include hydrodynamic interactions (drag minimization) (Geldart, 1987; Zelenko et al., 1996), inelastic grain-grain collision (collisional cooling (Lu et al., 2005; Wang et al., 2009) and viscous dissipation (Subbarao, 1986; Shuyan et al., 2008)), electrostatic forces, capillary bridging or van der Waals forces (Israelachvili, 1992; Podczeck, 1998; Visser, 1989).

Royer et al. (2009) investigated some of these effects in granular streams of particles freely falling from a small opening at the bottom of a hopper. For particles in the 50–150 micron range they tracked cluster formation with a high-speed video camera falling alongside the stream. They found that the glass beads developed clusters during a 2.5 meter free fall. These cluster formations were observed in an atmosphere or vacuum suggesting that hydrodynamic interactions may not be a cause. When a similar experiment was performed with 100-micron copper particles, clustering was not observed. If inelasticity was a driving force for clustering, clustering should have been more pronounced as copper has a coefficient of restitution of 0.9 compared to soda glass with a coefficient of 0.97. This together with the fact that the glass particle clusters did not slowly disintegrate during freefall suggested the presence of small cohesive forces in the nano-Newton range.

Clustering in this scenario results when impacting particles cannot escape from the energy well created by attractive forces. In the freefall experiments, the relevant region is right underneath the hopper opening, where the granular kinetic temperature has dropped to a level that clusters are no longer sheared apart, but the collision frequency is still high enough to lead to aggregation. Here, the lighter glass particles did not have enough impact energy to escape after a collision, while the heavier copper...
particles. Waitukaitis et al. (2013) performed experiments to become important with other types of insulating particles did.

nanoparticles to the surface of the glass beads. The addition of Aerosil silica nanoparticles to the surface of the glass beads. This resulted in the treated glass beads exhibiting a much reduced adhesion force in the AFM measurements and no longer showing particle clustering in the freefall experiments.

For the glass and metal particles in the experiments by Royer et al., short-range cohesion dominated over longer-ranged charging effects. Electrostatic charging can, of course, become important with other types of insulating particle material. Waitukaitis et al. (2013) performed experiments with freely falling streams of 200–300 micron zirconium dioxide silicate particles, in which they incorporated a Faraday cup at the bottom and had parallel electrodes along the particle free fall. By applying a horizontal electric field, the charge on individual particles could be extracted. Remarkably, while the particle stream as a whole was found to be essentially uncharged, the distribution of charge on individual particles showed broad tails around zero, indicating the existence of particles with large net positive or negative charges, equivalent in magnitude to the charge of several million electrons. It was noted that in these experiments the particle density in the freefall region was adjusted to be low (by using a smaller opening at the bottom of the hopper than Royer et al. used), such that particle-particle collisions were rare. Thus, the particle tracking during freefall was essentially a diagnostic of the triboelectric or contact charging that had occurred during earlier particle-particle contacts, most likely inside the hopper during the outflow process.

Waitukaitis et al. (2013; 2014) were able to show that such contact charging among particles made from the same material involves the transfer of negative charge and that the charging magnitude can be linked directly to the dispersion in particle size, as proposed earlier by Lacks et al. (2007; 2008). Particles smaller than the mean size tend to become charged negatively, while larger particles tend to acquire positive charge. However, at least for zirconium dioxide silicate particles Waitukaitis et al. found that the charge carrier could not have been electrons. The density of electrons in high energy trapped states at the particle surfaces was shown from thermoluminescence measurements to be at least four orders of magnitude too small than needed to account for the observed charging of these particles.

This suggested that the charge carrier responsible for triboelectric contact charging might be ions, such as OH- ions from the presence of water on the surface of the particles. Pence et al. (1994) and McCarty et al. (2008) have noted the importance of water, and with it OH- ions, on the surface of particles for contact charging. Unless one prepares surfaces under ultra-high vacuum conditions, thin water layers are generally unavoidable. Indeed, even in a vacuum at 10^-6 Torr, a molecularly thin layer of water adsorbs in one second, which is the definition of a Langmuir. However, we caution that to date there is not yet consensus about the precise tribocharging mechanism applicable to granular materials. For example, Baytekin et al. (2011) showed that contact charging of insulating materials can also occur when tiny, nano-sized chunks of solid surface material are transferred during contact.

The surface morphology of particles therefore appears to play a role on several levels as far as clustering is concerned: Asperities may increase the distance two particles can come into proximity, which controls the strength of short-ranged van der Waals and/or capillary forces. In addition, the surface morphology is likely to affect the efficiency of charge transfer during contact, which in turn affects long-ranged electrostatic interactions.

Lee et al. (2015) used high-speed video of 300 micron, free-falling zirconium dioxide silicate particles to explore how electrostatic charging increases the efficiency of particle capture and aggregation processes. They were able to track in detail how particles attract each other over distances of 100’s of microns, undergo multiple collisions, and finally stick to each other. Lee et al. were able to model the trajectories between successive collision events as Kepler-like orbits, taking into account the mutual polarization of the particles.

Lee’s et al. findings also showed that particles lost some translational kinetic energy upon collision, while particle rotation can have a significant effect. They were able to capture two particles having an electrostatic attraction for each other but clustering failed resulting from the rotation of one of those particles having a less than spherical shape. Asperities from a rough surface could have a similar effect (Wilhelm and Kwauk, 1948).

Lee et al. (2015) also noted that cluster formation often requires multiple bounces to dissipate the kinetic energy such that cohesion can dominate. Clustering was at its lowest probability for the collision of two particles. Collisions of a particle with a pre-existing cluster appeared to be more likely to lead to aggregation, owing to the ability of a cluster to deform internally. In other words, clusters have a lower coefficient of restitution than individual particles.

Thus, in the presence of electrostatic forces, particle clustering appears to be a two-step process. Coulombic...
forces between oppositely charge particles or forces arising from polarization of a less-charged particle by a highly charged particle, provide a long-ranged attraction. We note that polarization forces are always attractive, irrespective of the charge polarity on the participating particles. For single-peaked particle size distributions, where the peak of the associated charge distribution from collisions will be near zero charge (Waitukaitis et al. 2014), this means that clusters will tend to be comprised of (near) neutral particles that have aggregated around highly charged ones.

The second step in clustering involves short-ranged cohesive forces. Surface roughness and asperities can lead to more rotational momentum upon collision [Geldart and Wong, 1987; Cocco et al., 2010]. Fines may be less prone to this, as these particles have both less translational and rotational momentum which may also explain why clustering is more evident for many fluidized bed systems with particles smaller than 44 microns.

Longer-ranged electrostatic interactions make it possible to capture and aggregate particles even if they are not undergoing collisions that transfer rotational momentum to translational or by some other similar process. By preventing particle escape during multiple bounces, these interactions also lead to efficient capture when the impact energy would be too large to be sufficiently dissipated during a single collision. This greatly increases the clustering propensity. On the other hand, as clusters grow they eventually reach a size, where, depending on the cohesive strength and the background pressure, they will become large enough that other factors become controlling such as particle shear and drag.

5. Clusters and bulk shear forces

To discern the extent of particle clustering, flow measurements were taken in a 2.5-cm diameter fluidized bed using Freeman Technology’s FT4 Powder Rheometer in accordance to ASTM D7891 (2009). If particle clustering is pervasive throughout the emulsion, it should be detectable with the energy required to shear the fluidized bed. If particle clustering is restricted to the region around or below the bubbles, such detection should be limited to the number of bubbles. At low superficial gas velocities near the minimum bubbling velocity, this number should be low and the impact to shear the bed minimal.

The FT4 Powder Rheometer measures the resistance that a powder exerts on a rotating blade as it moves axially through a powder sample which is packed, fluidized or compacted. This resistance, measured as torque and axial force, quantifies the energy needed to rotate the blade. In short, the FT4 provides a measure of the stresses involved when the powder shears at different axial positions for a packed, compacted or fluidized bed. If clustering is prolific throughout the emulsion of a fluidized bed, then more energy would be needed to shear the solids than for a case where clustering is not significant. Similarly, if clustering is restricted to the region around or below the bubbles, the additional shear should be diluted by the rest of the non-clustering emulsion.

For this study, three samples were examined which consisted of FCC catalyst powder with varying levels of fines concentration (< 44 microns) at 3 % fines (low fines), 12 % fines (medium fines) and 100 % fines (high fines). Analysis consisted of filling a 5-cm diameter cylinder with 25 ml of material. Testing was done at room conditions and replicated for all data points. Testing consisted of measuring the energy needed for rotation in a packed and fluidized bed for up to five times the minimum fluidization velocity.

Fig. 9 shows the results from the permeability testing. As expected for flow through a packed bed, the pressure drop owed to the permeability was five times higher for the high fines case compared to the low or medium fines cases. This level of pressure drop can be typical of cohesive materials which is in agreement with Figs. 4 and 5 with the comparison of entrainment for Geldart Group A and B materials.

It is interesting that the low and medium fines case had similar permeabilities, with the low fines case showing only a slightly lower pressure drop relative to the medium fines case. Fig. 10 better illustrates this subtle difference. At low normal stresses (little compaction) this difference is more significant, but as the normal stresses are increased (higher compaction) this difference becomes small. The medium fines case appeared to be insensitive to increases in normal stresses from 2 to 14 kPa, suggesting fines provided a close packed bed upon filling.

This difference is more evident in Fig. 11 where 15 kPa of normal force on top of a packed bed of particles resulted in a certain level of compaction. The medium fines
case resulted in the least amount of compaction which is in agreement with Fig. 10. The less availability of fines results in less of a close pack situation. The high fines case showed the most compaction which is likely the result of weak cohesive forces preventing the bed from approaching a close packed condition with no normal force on the bed. Once force is applied, the cohesive forces are quickly overwhelmed with the applied force.

Consolidation from vibration at a constant amplitude and frequency provided a different trend. The low and medium fines case resulted in comparable consolidation indices (the degree of compaction with mechanical agitation) whereas the level of compaction for the high fines case was three times higher, as shown in Fig. 12. This difference suggests that even mild perturbations to the bed can break up the cohesive forces. Thus, if clustering is significant in the bulk, it appears to provide a lower bed density in the non-consolidated state. However, even low levels of energy in the environment seem to break up or reconfigure these clusters.

The most discernible trend with regard to the level of fines was observed with the Basic Flowability Energy (BFE) test. BFE is the energy required to establish a particular flow pattern in a conditioned, precise volume of powder. This flow pattern is a downward anti-clockwise motion of the blade, generating a compressive, relatively high stress flow mode in a packed bed of powder. The BFE is calculated from the work done in moving the blade through the powder from the top of the vessel to the bottom (i.e. during the downward traverse).

Fig. 13 shows the BFE for the three FCC samples in a packed bed state. The low fines case required the most energy needed for displacing powder. That level of energy decreased as the level of fines concentration increased in the samples suggesting the flowability of the bed becomes easier with increasing fines level. These results are consistent with the increasing formation of clusters with higher fines and resulting in more consolidation during vibration as shown Figs. 11 and 12. The granular stresses are less apparent with increasing fines levels. Presumably the higher fines levels provide larger or more clusters which owing to their weak cohesive forces act as a lubricant when under stress.

Indeed, these results suggest that the BFE might provide a trend that can be calibrated to the level of fines concentration which may be an indication of the level of particle clustering in terms of number and/or size. How-
ever, the work of Royer et al. (2009) and Waitukaitis et al. (2013) clearly shows that material type and surface morphology have a significant role in the level of particle clustering. Though, the BFE might be able to trend with a cluster level, that trend is most likely to be different for each material with respect to size, surface chemistry, morphology, etc.

**Fig. 14** shows the Aerated Energy (AE, the energy measured on the insert when air is being passed through the powder at a specified velocity) needed for rotating the blade with increasing levels of superficial gas velocities. This allows the measurement of the AE from a packed bed to fluidized bed state. The minimum fluidization velocity is apparent with this test and is represented by the leveling off of the normalized total energy needed to rotate the blade (integrated with respect to axial penetration). For the low fines case, the minimum fluidization velocity appeared to be at 0.002 m/s, whereas the high and medium fines case were at 0.004 m/s which is consistent with experimental measurements based on ASTM 7743 (2008).

Above minimum fluidization velocities, the high fines case consistently resulted in more energy needed for the rotating blade with significant cohesive forces prohibiting uniform fluidization. The medium fines case resulted in lower energy requirements compared to the low fines case for superficial gas velocities below 0.01 m/s, as a result of lower permeability and moderate cohesion. At velocities greater than or equal to 0.01 m/s, the trend reversed and more energy was needed for the medium fines case compared to the less cohesive low fines case. However, this may be the result of bed becoming fully fluidized in the low fines case with the onset of bubbling. With higher fines levels, the minimum bubbling velocity increases (Geldart, 1986) and beyond the minimum bubbling velocity, no further improvements in flow properties are observed.

The results from the FT4 tests between the minimum fluidization and minimum bubbling velocities, suggest that clustering might impede the flow of particles in a homogenous or smoothed fluidized bed. Because bed densities at fluidization are much less than that of a packed bed, the shear forces may be less than the cohesive forces. Hence, the energy needed for rotating the blade increased as it would increase for increasing particle sizes.

From the results presented in **Figs. 9** through **14**, the shear stresses and compaction of a packed bed are influenced by the level of fines concentration. The BFE and AE tests show the best trend with respect to increasing fines concentration, supporting the assumption that clusters could be acting as a lubricant in granular flow of packed beds, yet limiting the extend of homogeneous dispersion in fluidized beds.

The AE test quantifies the energy requirements needed for rotating the blade in a fluidized bed. For measurements between the minimum fluidization and minimum bubbling velocities, higher fines levels appear to coincide as a trend with higher AE requirements. Since the bed is fluidized, the shear stresses associated with particle shape and surface roughness have less of an influence as in the BFE test where results predominantly depend on cohesive strength. Other factors such as particle shapes and structural integrity (i.e., attrition) should also be a consideration.

However, it is still circumstantial that the AE test trends with the level of clusters either by number or size. Additional data are needed to provide that trend either by directionally measuring cluster number and size in a bed under similar conditions and using it for correction to entrainment calculations, or comparing it directly to entrainment data.

The results of the FT4 Powder Rheometer testing suggest that cohesive forces presumably resulting in particle clustering are prolific in the bulk solids and are not confined in the region around or below a bubble. This is evident with the non-monotonic changes in permeability and compaction with increasing fines level where bubbles were not present. The AE testing also suggests particle clusters are not restricted to bubbles as more energy was needed with increasing fines levels below the minimum bubbling velocity under homogenous or smooth fluidization.

### 6. Conclusions

With particle clustering being linked to the morphological and electrostatic nature of the particle surface, entrainment rate calculations are highly material specific. Coupled with the reflux of particles or clusters in the freeboard because of internals such as cyclones and expanded heads, it appears unlikely that a fundamentally-based CFD model would be effective in determining the en-
Direct evidence of particle clustering in a fluidized bed has been observed. Particle clusters form in the bed and their size may be dependent on the residence time in the bed (i.e., bed height and gas velocity). The mechanism for clustering appears to be resulting from a combination of short-ranged cohesion due to van der Waals or capillary forces and longer-ranged electrostatic interactions, with particles becoming charged by transfer of electrons or, more likely, ions during contact. The small particles or fines provide an effective conduit in moving the charge throughout the fluidized bed. Fines have a higher mobility in fluidized beds. However, rotational movement of a particle could overpower the electrostatic forces, especially for the larger particles. Thus, particles with low sphericity or high surface roughness may be less likely to form particie clusters.

Despite the mechanism, microscopic particle clustering affects the fluidization behavior and has a significant effect on entrainment, a design parameter critical for fluidized bed scale-up. To achieve better accuracy with entrainment rate calculations and with the CFD modeling of entrainment, this clustering needs to be accounted for.

Preliminary testing using the FT4 Powder Rheometer for measuring the energy needed to shear FCC catalyst samples at varying levels of fines concentration in a packed and fluidized bed suggest fines play a role here as well. However, such testing under fluidized conditions relaxes the effects of other factors such as particle shape and surface roughness on the shear stresses. Furthermore, the results of this aeration test found a trend only with the level of fines concentration and not directly with the level of particle clusters which were unknown.

Nonetheless, particle clustering in a fluidized bed appears to be significant throughout the bed and not confined to the bubble, even though visual observations were restricted to the region around the bubble. Permeability, compaction, and unconsolidated shear stress measurements showed a good trend with the level of fines, a promoter of particle clustering, in a packed bed. Furthermore, the AE testing further confirmed this by showing a monotonic trend in energy requirements with increasing fines levels below the minimum bubbling velocity.

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