



Particle clusters in and above fluidized beds

Ray Cocco^{a,*}, Frank Shaffer^b, Roy Hays^a, S.B. Reddy Karri^a, Ted Knowlton^a

^a Particulate Solid Research, Inc., 4201 W 36th Street, Suite 200, Chicago, IL 60632, United States

^b National Energy Technology Laboratory, Department of Energy, Mail Stop 84-202, Cochran's Mill Road, Pittsburgh, PA 15236, United States

ARTICLE INFO

Article history:

Received 15 October 2009

Accepted 18 February 2010

Available online 2 April 2010

Keywords:

Fluidization

Entrainment

Elutriation

Particle clusters

High-speed video

ABSTRACT

High-speed video imaging of particle clusters in and above a fluidized bed suggests that clustering is significant for FCC catalyst and polyethylene powders. Based on fluidized bed experiments at varying fines concentration, bed heights and bed internals location, the dominant mechanism for clusters in the freeboard appears to be cluster formation in the bed. Some of these clusters are then subsequently ejected into the freeboard region. Hydrodynamics does not appear to be solely responsible for cluster formation. Cohesive forces such as electrostatics, capillary and van der Waals forces, appear to play a significant role in particle cluster formation. The proposed mechanism suggests that particle shear produces collisional cooling that allows the granular temperature to decay to where these cohesive forces can dominate. The decrease in the granular temperature appears to be dependent on the particle properties and surface morphology. Collisions that only redirect the particle or increase particle rotation, limits this reduction in the granular temperature such that cohesive forces are less of an impact. In the case of risers, where large shear streams are prevalent, these clusters and the corresponding drag forces may result in the formation of larger clusters or streamers.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

The clustering of particles in fluidized beds and circulating fluidized beds continues to be a fundamental issue in granular-fluid hydrodynamics. Wilhelm and Kwauk [1] were among the first to show evidence of particle clustering in fluidized beds. Others followed suit with similar experiments [2–10]. Kaye and Boardman [10] proposed that particle clustering becomes significant in many systems where solids concentrations exceed 0.05%. Jayaweera et al. [3] used particle drop experiments to show that clusters have stable size ranges of two to six particles. Clusters greater than six particles tend to split and form stable sub-groups of clusters. Fortes et al. [7] observed similar-sized clusters to be stable up to a particle Reynolds number of 1800.

Although neglected, the role of particle clusters with granular-fluid hydrodynamics can be significant. Yerushalmi et al. [11] first illustrated this with observations of slip velocity in fast fluidized beds that were significantly larger than expected for the particle size. This was later attributed to particle clusters [12]. Matsen [13] concurred with their findings and added that cluster size may be dependent on solid loadings. Geldart and Wong [14] observed the reduction of entrainment in a fluidized bed with the addition of Geldart Group C particles to a bed of Group A powder. They suggested that the Group C particles or fines may be adhering to the large Group A particles, or to each other. Regardless of the mechanism, the

outcome is the formation of particle clusters that have higher slip velocities and are less likely to be entrained. Similar findings were reported by Baeyens et al. [15] who noted that entrainment leveled off at some critical particle size between 25 and 40 μm where cohesive forces are exceeded by other forces. Interesting enough, this entrainment rate depression due to particle clustering was not dependent on particle density of the added polymer and metal fines to the bed of Geldart Group A powder. Choi et al. [16] also looked at fines and their effect on the elutriation of coarse particles. He found that the elutriation of fines was not affected by the particle size distribution in the bed. Li et al. [17] suggest that there may be a bridging effect with fine powders and that the elutriation rate constant of Group C or Group A particles is not only affected by the properties of the elutriated powders or particles and gas velocity, but also by both the weight fraction and size of the Group C powder in the bed.

Particle clusters have been found to be significant in the risers of circulating fluidized beds as well. O'Brien and Syamlal [18] concluded that particle clusters, on the order of ten particle diameters in size, must exist in risers to explain the higher than expected drag forces observed in the circulating fluidized bed experiments of Bader and coworkers [19]. Agrawal et al. [20] also had similar findings and provided a correction to the drag law to reflect on meso-scale features that must exist in riser flow. Guenther and Breault [21] used wavelet decomposition of backscattered light from a fiber optical probe to discern these particle clusters in a riser. Their results were in good agreement with O'Brien and Syamlal's calculations that clusters exist as formations of 10 to 20 particles. They also observed that the cluster size was relatively independent of the superficial gas velocity.

* Corresponding author.

E-mail address: ray.cocco@psrichicago.com (R. Cocco).

Horio and Kuroki [22] were able to use a laser sheet technique to visualize particle clusters in circulating fluidized beds. At low loadings, they found that the gas flow did not suspend the particles homogeneously and allowed for gas pockets. At higher loadings, the inhomogeneity translated into particle clusters. For 70- μm sized particles, they observed clusters on the scale of 5 to 20 mm, which is significantly larger than those observed in previous studies [3,7,18,21]. Noymer and Glicksman [23] used heat transfer measurements at a riser wall to determine that particle clusters were on the order of 10 mm in size. Rhodes et al. [24] observed particle swarms with cord lengths of 10 to 15 mm. Sharma et al. [25] used localized capacitance probes to examine the density within a cluster. In a fast fluidized bed, they found that cluster density was about 2.4 times the emulsion density.

It appears that in fluidized beds and circulating fluidized beds, particle clusters can exist on two scales: (i) micro-clusters consisting of a few to 20 or more particles, and (ii) macro-clusters consisting of hundreds of particles that may exist as spheres, strands or streamers. Which type of particle cluster exists may depend on operating conditions and material properties. In the case of circulating fluidized beds, both types of clusters may exist.

The underlying question with these clusters is what is the mechanism for their existence. This paper will attempt to address at least some of the mechanisms involved with micro-clusters. Based on evidence from pilot and commercial scale plants along with high-speed video of a cold-flow fluidized bed, the mechanism of particle clustering in and above fluidized beds is examined.

2. Background

Hays, et al. [26] presented several cases where evidence of particle clustering was observed in and above a fluidized bed. In the first case, they attempted to mimic a cyclone flooding problem observed in a commercial-scale fluidized plant. After a period of continuous operation, the entrainment rate increased beyond design specifications. This was duplicated in the lab with a 6-in. (15-cm) diameter fluidized column with a static bed height of 52 in. (132 cm) of the same Geldart Group A powder (d_{p50} of 55 to 60 μm) used in the commercial process. The test unit was operated in batch mode at a superficial gas velocity of 0.66 ft/s (0.2 m/s).

Fig. 1 shows the entrainment flux measured at the outlet of the unit and the fines weight fraction (defined as particle sizes smaller than 44 μm) for the bed and entrained material. Only after the fines concentration decreased in the bed did the entrainment rate increase by ten-fold. Further depletion of fines from the bed resulted in a subsequent drop in the entrainment rate. For the commercial unit, the flooding of the cyclone may have been related to a reduction of fines in

the bed, which resulted in a large increase in the entrainment rate. The cyclones were not designed to handle the presumed ten-fold increase in the entrainment rate.

These results are consistent with earlier studies of Geldart and Wong [14], Choi et al. [16] and Li et al. [17]. The addition or presence of fines in a fluidized bed of Geldart Group A powder resulted in a depression of the entrainment rate. Hays' study further confirmed the role of fines by showing that the removal of fines from a fluidized bed resulted in a significant increase in the entrainment rate, presumably resulting from a decrease in particle clusters.

PSRI observed similar results with coke powders of two different particle size distributions (d_{p50} of the coarse particles was about 76 μm and that of the fines was about 20 μm) in a 15-cm diameter fluidized bed. Table 1 shows a comparison of entrainment fluxes measured in the 15-cm diameter fluid bed at two different fluidizing gas velocities of 0.3 and 0.46 m/s for both the coarse and finer coke particles. At 0.3 m/s, the entrainment flux of 76 μm coke was about 16 times that of the 20 μm coke particles, suggesting the 20 μm particles were clustering. However, increasing the gas velocity from 0.3 ft/s to 0.46 m/s caused the entrainment flux of the finer coke particles to increase by 184 times. This was similar to the entrainment flux for the 76 μm coke particles and suggests the higher velocities limit cluster growth or stability.

In the second case, Hays et al. [26] examined the batch fluidization of FCC catalyst fines (d_{p50} of 27 μm) in a 6-in. (15-cm) diameter fluidized bed equipped with a cyclone, dipleg and L-valve particle recovery system. Experiments were performed at a superficial gas velocity of 1.8 ft/s (0.56 m/s). At this velocity, all of the material in the fluidized bed can be considered as entrainable. As expected, the bed height decreased slowly with time due to the entrainment of material from the bed some of which was not collected by the cyclone. However, the entrainment rate increased with time. In order to determine if it was the decrease in bed height that was causing the increase in entrainment, material was removed from the dense phase region of the bed while it was fluidized after 4500 s. As shown in Fig. 2, the removal of bed material (vertical dashed line) from the column caused a significant increase in the entrainment rate. Hays' data suggest that the entrainment rate was inversely proportional to the bed height, at least for beds with high fines concentrations. This conflicts with the expectation that at greater disengaging heights (lower bed heights) result in lower entrainment rates.

The effect of bed height was further explored by measuring the entrainment rate for the FCC catalyst fines at various bed heights. In separate fluidization experiments, the entrainment rate at six different bed heights was explored. As shown in Fig. 3, an inverse linear relationship was observed between the fluidized bed height and the corresponding entrainment rate. The opposite was expected since an increased bed height raises the transport disengagement height. If the outlet is not high enough, the higher bed height results in a higher entrainment rate. The reduction of entrainment with increasing bed height suggests that the size of the particle clusters is dependent on the bed height.

The third case presented by Hays et al. was with FCC catalyst powder (d_{p50} of 70 μm) with 5% fines in a 6-in. (15-cm) fluidized bed with and without baffles located at 1.6 and 2.5 ft (0.5 and 0.75 m) above the distributor plate. The baffles resembled simple grating

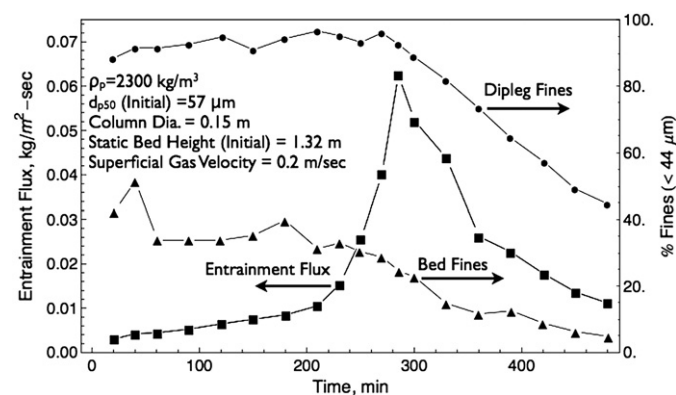


Fig. 1. Elutriation of fines from a 6-in. (15-cm) diameter fluidized bed at 0.66 ft/s (0.2 m/s) superficial gas velocity. Fines level and solids entrainment rate are tracked with time.

Table 1

Entrainment flux for flexicoke particles in a 0.15-cm diameter fluidized bed for two different particle size distributions at two different superficial gas velocities.

Particle size	Entrainment flux at specified superficial gas velocity	
	0.30 m/s	0.46 m/s
20 μm	0.002 kg/m ² s	0.355 kg/m ² s
76 μm	0.035 kg/m ² s	0.448 kg/m ² s

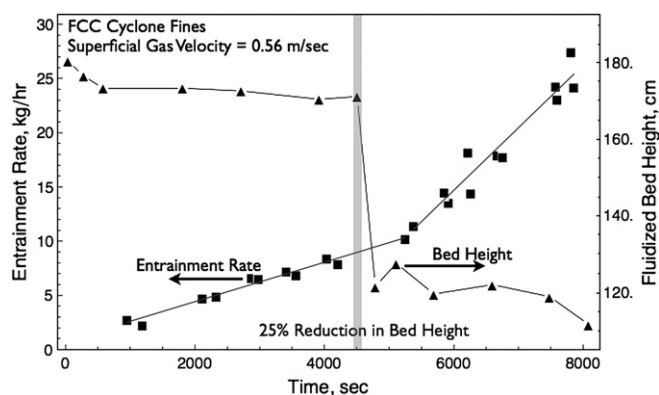


Fig. 2. Effects of entrainment rate from a bed of FCC catalyst fines when the bed height was decreased by 25% at 4500 s. Superficial gas velocity was at 1.8 ft/s (0.56 m/s) at room conditions.

commonly used for floor decking. The bed height was at 2.5 ft (0.76 m). As shown in Fig. 4, the presences of baffles resulted in an increase in the entrainment flux at higher gas velocities.

In a similar experiment with coke (d_{p50} of 150 μm) having no fines, this increase in entrainment rate in the presence of baffles was not observed, as shown in Fig. 5. Unlike the Geldart Group A material, the presence of eggcrate type baffles did not increase the entrainment flux. These data suggest that the particle clusters, possibly responsible for the entrainment rate depression, may be formed in the bed region. In other words, particle clusters may be formed in the fluidized bed and when ejected into the freeboard, have a higher slip velocity than single particles with the potential to fail to be entrained. The deeper the bed, the larger or more stable the particle cluster.

However, this hypothesis is based on circumstantial evidence. To date, no direct evidence of particle clusters existing in the fluidized bed has been presented. In order to provide more conclusive evidence of the nature of particle clusters, a special optical system was developed and used to examine particle cluster behavior in and above a fluidized bed. The results of this study are presented below.

3. Experimental

3.1. Materials

For this study, two materials were used: polyethylene and FCC catalyst powders. The polyethylene powder is commonly used for coatings and listed under the name of Plascoat™ PPA 571 (Rocky Mountain Salvage). The FCC catalyst powder was material obtained from a commercial FCC unit and is commonly called equilibrium FCC catalyst powder (Rocky Mountain Salvage). Both materials are con-

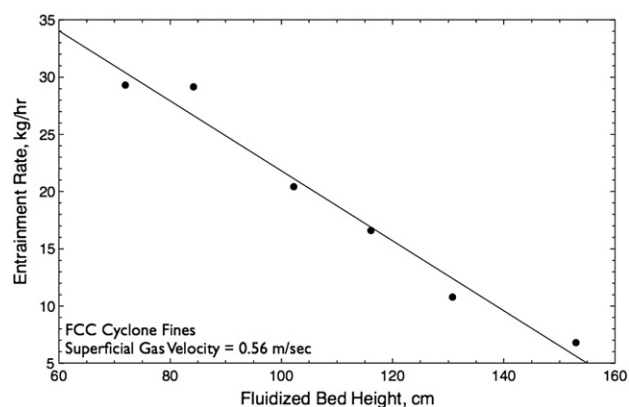


Fig. 3. Entrainment rate versus bed height from a bed of FCC catalyst fines at a superficial gas velocity of 1.8 ft/s (0.56 m/s).

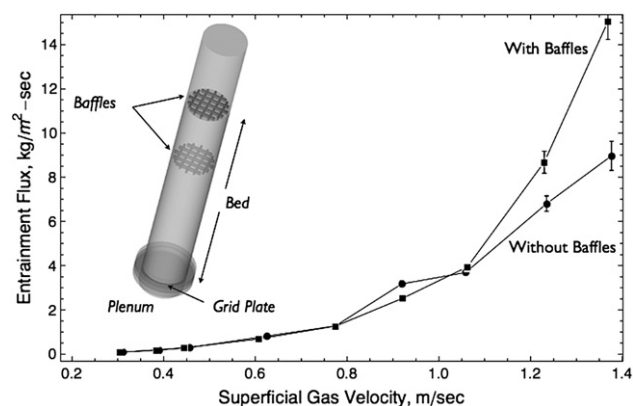


Fig. 4. Entrainment flux from a 6-in. (15-cm) diameter bed of FCC catalyst with and without baffles at various superficial gas velocities.

sidered to be Geldart Group A powders with mean particle sizes of 70 and 76 μm , respectively. The particle size distributions for each material are presented in Fig. 6. The particle densities for the polyethylene and FCC catalyst powders were 400 and 1500 kg/m^3 , respectively.

3.2. Fluidized bed

Experiments were carried out in a 6-in. (15-cm) diameter by 15-ft (4.6-m) high unit constructed of acrylic tubing. The distributor consisted of a perforated plate with 61 holes with a diameter of 3/16 in. (0.47 cm). Filter cloth was positioned below the perforated plate to prevent solids seepage into the plenum region. The outlet consisted of a 2-in. (5-cm) diameter port on the top of the unit. A three-stage cyclone system was used to collect entrained material.

Two 0.5-in. (1.2-cm) diameter ports were added to the unit to allow access for the high-speed video imaging. One port was located 6 in. (15 cm) above the perforated plate. The other port was located 8 in. (20 cm) below the top of the unit. Compressed air at room temperature was used as the fluidizing gas.

3.3. High-speed video

High-speed video images were obtained using a Vision Research Phantom v7.2 camera. Although the camera is capable of 75,000 fps, only frame rates ranging from 3000 to 6000 fps were used. The camera was fitted with a C-mount fitting to connect to the boroscope that was inserted into the fluidized bed.

Vision Research Phantom Camera Control Software (version 9) was used for image downloading. Particle size analysis of the high-speed video images was conducted using ImageJ software (<http://>

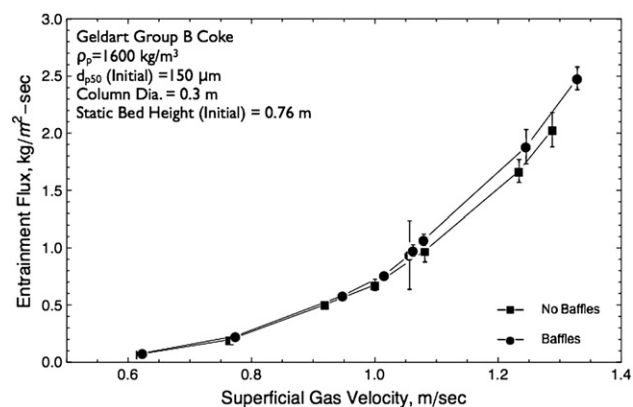


Fig. 5. Entrainment flux from a 12-in. (30-cm) diameter bed of fluid coke powder with and without baffles at various superficial gas velocities.

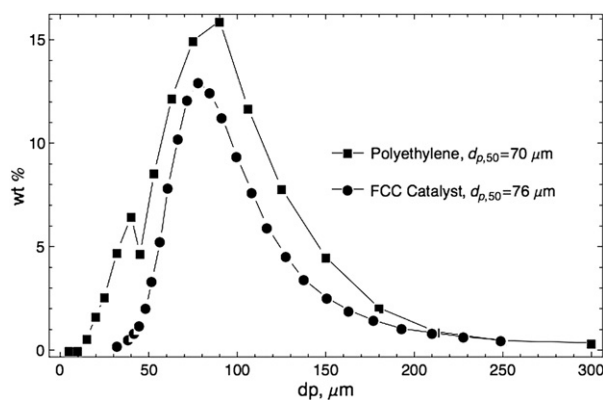


Fig. 6. Particle size distributions for Plascoat™ 571 polyethylene and FCC catalyst powder.

rsbweb.nih.gov/ij/) using a Waterfall filter. Particle tracking was conducted with the use of proprietary software from the National Energy Technology Laboratory (NETL).

3.4. Optical probe

An Olympus R100-038-000-50 Industrial Rigid Borescope was modified to allow imaging of particles and clusters in the freeboard and in the bed. This borescope has a depth of field of 5 mm to infinity. The borescope was fitted with a 6-mm diameter optical spacer (Melles Griot) to account for the distance between the borescope face and the focal length. This prevented particles closer than the focal length from blurring the images or reducing the lighting for the imaged particles. The spacer was further secured using a stainless steel guard collar to protect the instrument when in the fluidized bed.

The Olympus borescope allows for internal lighting. A xenon light source with an Olympus Liquid-Filled Light Guide was used to supply lighting through the borescope probe. External lighting was used whenever possible.

4. Results

4.1. Polyethylene

Fig. 7 shows a series of images taken at the center of the fluidized bed in the freeboard region (8 in. (20 cm) from top of unit) using the Phantom camera and borescope arrangement. Particle tracking has been overlaid over each image. Although the polyethylene powder

had a mean particle size of 70 μm, clusters on the order of 100 to 500 μm and larger were readily observed. Since the modified borescope arrangement was used, wall effects were minimal.

The size of the clusters was obtained from 100 different images from the high-speed video. Fig. 8 shows the particle size distribution of the entrained material obtained from image analysis. Fig. 8 also shows the particle size distribution of the bed using a Microtrac™ laser diffraction analyzer. The mean cluster size was measured to be 175 μm. However, this distribution also includes particles not part of any clusters. The actual cluster size distribution can be obtained by deconvoluting the particle component from the high-speed video curve shown in Fig. 8. If a log normal distribution is assumed along with constraints that the particle component does not move peak position or peak width at half maximum, then the cluster size distribution can be deconvoluted from the high-speed video curve in Fig. 8.

The result of that deconvolution process is shown in Fig. 9 and suggests that the cluster size distribution ranges up to 700 μm with a mean cluster size of 240 μm. Assuming both particles and clusters are spherical and clusters are arranged in a close packed configuration with a void fraction of 0.35, the mean cluster size of 240 μm suggests that the number of particles per cluster is on the order of 26 particles. A ratio of the presumed particle peak and the deconvoluted peak in Fig. 9 suggests that 75% of the material observed in the freeboard are clusters with the remaining 25% being single particles. Details of the measurements are presented in Table 2.

A similar procedure was performed on the particles in the bed region. The modified borescope was installed in a second port located 6 in. (15 cm) above the perforated distributor plate. The borescope was extended to the center of the bed, about 3 in. (8 cm) from the wall. Fig. 10 shows several frames obtained from the high-speed imaging. Some material did stick to the probe face distorting some of the images. Presumably this is a result of electrostatics even though Larostat 519 (BASF) was added to the bed material prior to experimentation. However, moving clusters were observable with the video imaging. The cluster size range appeared to be similar to that observed in the freeboard, although detailed analysis similar to that conducted for the freeboard material was not possible because of the distortions. Regardless of these difficulties, high-speed imaging in the bed region above the distributor and away from the wall showed that large particle clusters can exist in a fluidized bed.

4.2. FCC catalyst

The modified borescope and Phantom camera arrangement was also used in a fluidized bed containing FCC catalyst. The borescope

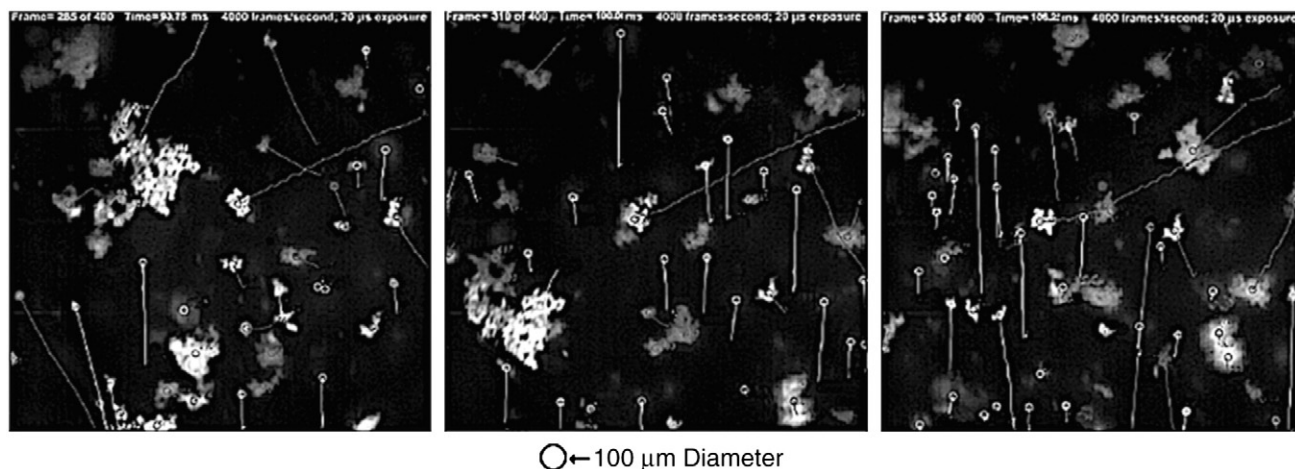


Fig. 7. Several consecutive frames (in increments of 35 frames) of Plascoat™ 571 polyethylene in the freeboard region of a 6-in. (16-cm) diameter fluidized bed at a superficial gas velocity of 2 ft/s (0.61 m/s). Images were collected at 4000 fps with a 20 μs exposure time.

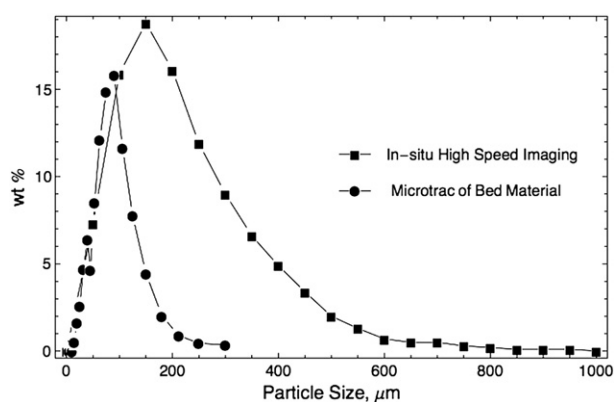


Fig. 8. Particle size distributions of bed material (from a Microtrac) and of freeboard material from high-speed video analysis.

was first inserted in the top port at the center of the freeboard. Fig. 11 shows several frames of the video obtained for particles and clusters in the freeboard region. As with the polyethylene powders, the FCC catalyst tended to cluster in the freeboard region. A statistical analysis of this video suggested that 30% of the material in the freeboard existed as particle clusters having an average size of 11 ± 5.0 particles. Thus, the FCC catalyst powder appears to exist as clusters in the freeboard, but cluster concentration was less than half the cluster concentration of the polyethylene powder. In addition, clusters appeared to be only about half the size of the clusters for the polyethylene powder.

As with the polyethylene study, the boroscope and Phantom camera arrangement was inserted into the bottom of the fluidized bed of FCC catalyst powder. The boroscope was positioned at the center of the bed to insure that wall effects were not an issue. Consecutive images from the collected video in the fluidized bed of FCC catalyst powder are shown in Fig. 12. Larger and more clusters were observed in the bed than in the freeboard. A statistical analysis of the video obtained in the fluidized bed suggested that 41% of FCC catalyst material existed as clusters with an average cluster size of 21 ± 1.7 particles.

Because of the bed density, cluster imaging was conducted during the period when a gas bubble passed the face of the boroscope. The clusters shown in Fig. 12 are in a much lower solids concentration than that of the emulsion phase. 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.

Fig. 13 shows three consecutive images where the video was magnified about five times that for Fig. 12. The clusters shown in Fig. 12 are more definitive, and show that the clusters exist as both

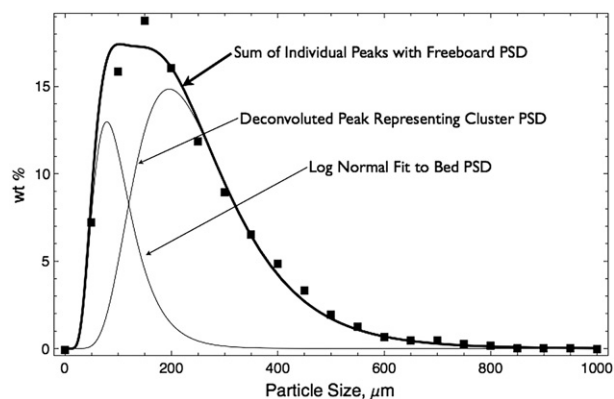


Fig. 9. Deconvolution of distribution representing freeboard material into particle and cluster size distributions.

Table 2

Median and Sauter mean particle sizes for bed material, freeboard material and deconvoluted clusters.

PSD analysis	d_{p50} , μm	$d_{p\text{Sauter}}$, μm
Bed material via Microtrac	70	63
Clusters (deconvolution)	240	212
Total (high-speed video)	175	157

large particles with fines, or as a large number of fines. Rarely was a cluster of only large particles observed. In some cases, large strands of ten or more particles were observed consisting of both large and small particles.

5. Discussion

5.1. Cluster measurements

Using an achromatic lens boroscope or rigid boroscope fitted with an optical glass spacer to extend the probe to the focal length, particle clusters were observed and measured in and above a fluidized bed. Only after using a xenon light source with a liquid-filled light guide (connected to the boroscope) along with sufficient external lighting was the resolution high enough to identify, size and track the particle clusters. Fiber optic boroscopes were insufficient in providing the resolution needed for these sizes of particles and clusters. With the addition of a high-speed camera, the particle size, particle trajectory, cluster size, and cluster trajectory can be obtained, provided a statistically significant amount of frames are analyzed.

5.2. Cluster behavior in and above a fluidized bed

Using the modified boroscope with a Phantom camera, particle clusters were readily observed in the freeboard region, as shown in Fig. 9 for polyethylene powder and Fig. 11 for FCC catalyst powder. For polyethylene powder, the average cluster size was $240 \mu\text{m}$ corresponding to approximately 29 particles, but clusters as large as $700 \mu\text{m}$ were measured. Assuming a packing fraction of 0.65, this corresponds to clusters containing 650 particles. Based on the probe measurements, more than 75% of particles in the freeboard were particle clusters. Thus, this phenomenon is not only evident, it is prevalent. In all cases, the clusters were not spherical or rigid.

Similar findings were found for the FCC catalyst particles although smaller and fewer clusters were observed. Similarly, Jayaweera et al. [3] and Fortes et al. [7] reported particle clustering for FCC catalyst material, but with smaller-sized clusters than observed in this study. Both the polyethylene and FCC catalyst powders had a similar particle size distribution as shown in Figs. 5 and 6. Thus, clustering appears to not only be dependent on the fines level but the material itself. Particle density may also be a factor, but previous studies by Baeyens et al. [15] suggest otherwise, at least for added fines.

Both Figs. 9 and 11 reveal why lower than expected entrainment rates were observed for both these materials. As expected and postulated by Hays [26], Geldart and Wong [14] and Choi et al. [16], particle clustering results in particle sizes too large for the drag force to carry the particles sizes out of the unit. Thus, fines that would have been easily entrained out of the unit now fall back to the fluidized bed. However, how these clusters form and the underlying mechanism for the cohesion still needs to be addressed.

Figs. 10, 12, and 13 show particle clusters in the fluidized bed near the grid plate for both polyethylene and FCC catalyst powders. Although Figs. 1–5 suggest particle clustering occurs in the fluidized bed, evidence was circumstantial. These images clearly show that particle clusters exist in the fluidized bed, even near the distributor. However, it is uncertain if these clusters occur only in the bubble region (cloud phase) or in the emulsion phase as well. Particle

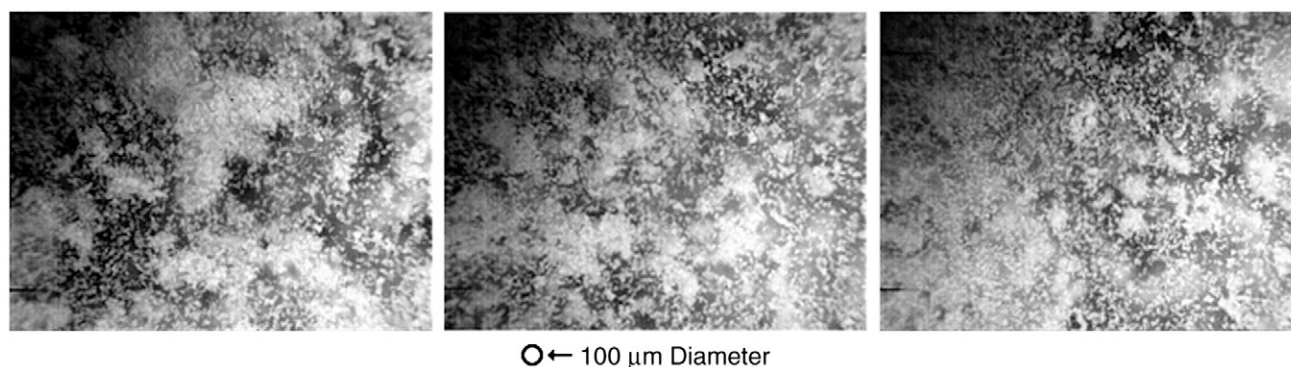


Fig. 10. Selective frames of Plascoat™ 571 polyethylene in the bed region of a 6-in. (16-cm) diameter fluidized bed at a superficial gas velocity of 2 ft/s (0.61 m/s). Images were collected at 4000 fps with a 20 μ s exposure time.

densities were too high to discern clusters when looking at the emulsion region. Only with the occurrence of a bubble near the probe were particle clusters observed. Thus, for Geldart Group A powders with fines, particle clustering does occur in fluidized beds either in the emulsion and bubble regions or just the bubble regions. Subbarao [27] originally postulated that there was a relationship between bubbles and cluster formation. Perhaps the bubble serves as a concentrator of fines, which better promotes clustering. Another explanation could be that the bubbles serve as a concentrator of clusters that were initially made in the emulsion phase.

Results also suggest that particle clusters form in the fluidized bed and get ejected into the freeboard instead of clusters forming only in the freeboard region. Particle clustering most likely does happen in the freeboard, but it may not be the dominating contributor to particle clusters in the freeboard. Kaye and Boardman [10] noted that loadings greater than 0.05% are needed for clustering to occur. Loadings may be too low in the freeboard region, especially at the top of the bed, for sufficient particle concentrations to promote particle clustering. Fig. 4 also supports the theory that particle clusters occur in the fluidized bed and then get ejected into the freeboard. The addition of baffles in the bed and near the top of the bed resulted in higher entrainment rates suggesting baffles break up the clusters before being ejected into the freeboard. However, this only occurred at high superficial gas velocities. Below superficial gas velocities of 1 to 1.2 m/s, the clusters appear to remain intact, suggesting insufficient shear was available to break up the clusters. Conducting the same experiment with larger coke powder showed no difference in entrainment rates with respect to the baffled and unbaffled cases, as shown in Fig. 5. The coke

material had fewer fines available for cluster formation. However, coke clustering for smaller particles is possible as indicated in Table 1.

Fig. 3 suggests that the height of the fluidized bed may control the cluster size or frequency. As the bed height was increased for the FCC catalyst material, entrainment rates decreased. This result suggests that formation of large clusters may not occur instantaneously, and sufficient time in the emulsion or bubble region is needed for large particle clusters to form. Thus, a particle cluster may form near the bottom of the bed and continue to grow as it migrates to the top of the bed, possibly with the help of bubbles. At the top of the bed it is either entrained or circulates back down to the bottom of the bed. Several cycles of the circulation may be needed to build large clusters. As bed height is increased, this large circulation zone becomes more dominant and the possible residence time of a particle cluster in the bed becomes extended. Fig. 14 provides an illustration of this proposed mechanism.

The results from using baffles in a fluidized bed, as shown in Figs. 4 and 5, also support this mechanism. The addition of baffles may not only serve as a mechanism to break up clusters (especially at low superficial gas velocities), but to disrupt the larger recirculation zones in the fluidized bed and reduce the time a cluster spends in this cycle. Instead of one large recirculation zone, several recirculation zones may develop with only the top recirculation zone contributing to the particle clusters that can be entrained, as illustrated in Fig. 14.

5.3. Particle cohesive forces

Even though there is sufficient evidence of particle clustering, the underlying mechanism for this clustering is still unclear. For Geldart

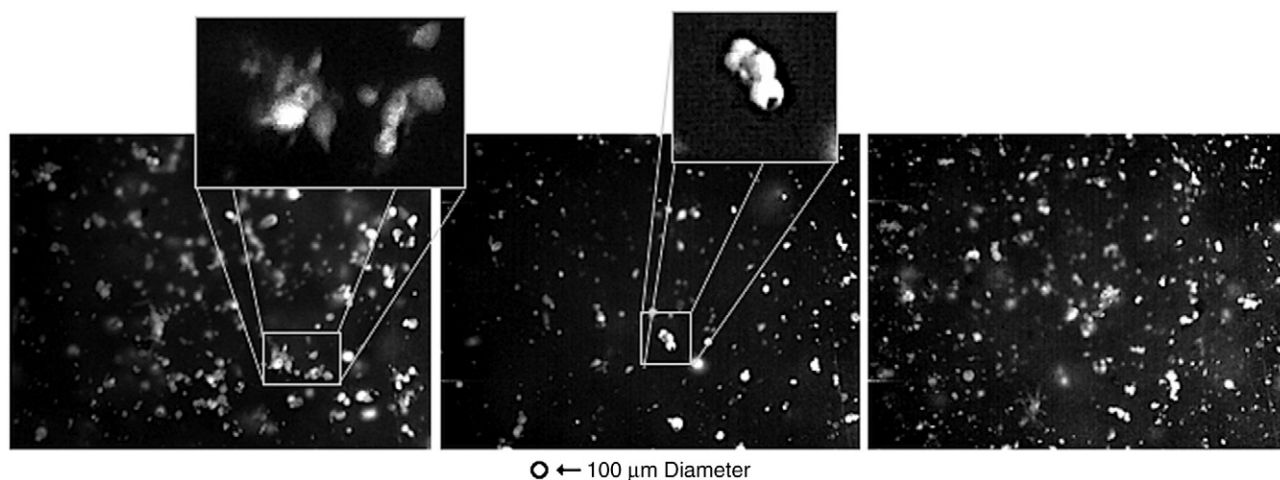


Fig. 11. Selective frames of FCC catalyst in the freeboard region of a 6-in. (16-cm) diameter fluidized bed at a superficial gas velocity of 2 ft/s (0.61 m/s). Images were collected at 4000 fps with a 20 μ s exposure time.

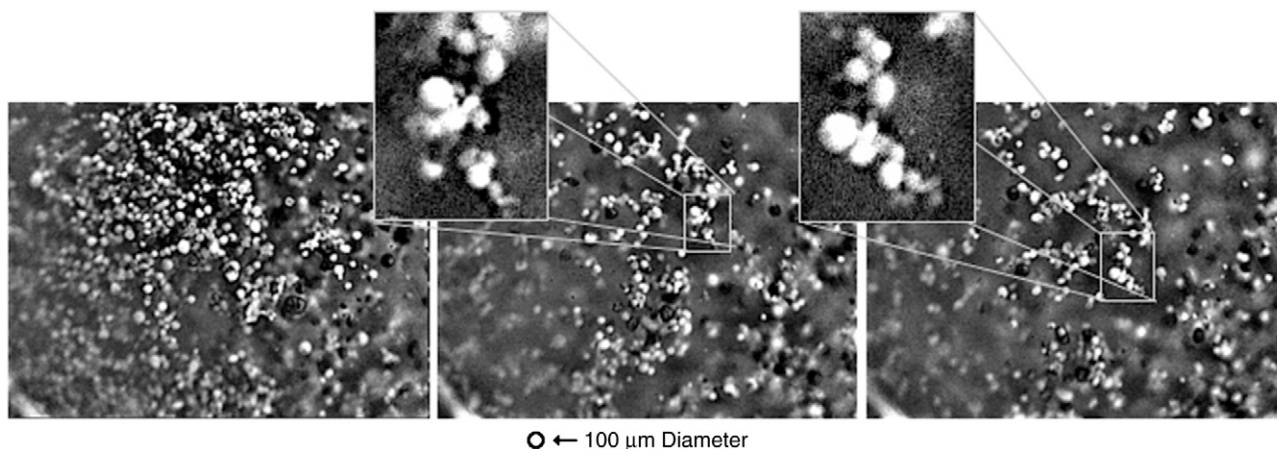


Fig. 12. Several consecutive frames of FCC catalyst in the bed region of a 6-in. (16-cm) diameter fluidized bed at a superficial gas velocity of 2 ft/s (0.61 m/s). Images were collected at 4000 fps with a 20 μ s exposure time.

Group A powders, the forces or possible interactions responsible for particle clustering include; hydrodynamics (drag minimization), inelastic particle collisions (collisional cooling or dissipation), electrostatic charging (coulombic), capillary, and van der Waals [28]. However, no one force may be responsible for all types of particle clustering. It is more likely that several forces may play roles of varying magnitudes depending on the particles and the environment.

Hydrodynamic interactions with a particle may certainly have a role. Horio and Clift [29] noted that particle clusters, “a group of loosely held together particles”, are the result of hydrodynamic effects. Squires and Eaton [30] proposed that clustering was due to turbulence modification from an isotropic turbulent flow field, which is an expansion of the work of Wylie and Koch [31]. Subbaroa [27] proposed that a uniform dispersion of particles in a gas is an unstable state and that particle clustering was inherent to the hydrodynamics of bubbles. The nature of this study was only able to confirm the presence of such clusters in and near the bubble region, but it did not provide evidence that clusters exist elsewhere in the bed. All of these studies proposed that viscous dissipation of the gas phase promotes the formation of particle clusters whether due to drag or turbulence reduction.

However, the presence of particle clusters in the fluidized bed suggests such dissipation may not be the dominant mechanism. A study conducted by Royer et al. [32] suggested that particle cluster formation, such as that observed in this study, may not be due to hydrodynamics. Royer and coworkers did a series of powder drop experiments using glass beads with an average particle size of 110 μ m. Using a high-speed camera falling in sync with the powder, Royer was able to show that particles cluster in a fashion similar to a stream of

liquid breaking up into droplets due to surface tension. Their study also showed that particle clusters formed whether the absolute system pressure was 101,325 (ambient) or 300 Pa (vacuum). In fact, better-formed clusters were observed at the lower pressure due to the fewer particles shedding off from the clusters due to drag. Little change in clustering behavior was observed for the glass beads in an electric field.

Royer and coworkers [32] expanded on this study to include powder drop experiments of copper powder and silver-coated glass beads. As with the standard glass beads, the silver-coated glass beads were also reported to cluster. Yet, copper powder did not show any evidence of particle clustering in the powder drop experiment. Copper has a lower coefficient of restitution than glass (0.9 for copper versus 0.97 for glass [33]). Thus, cluster formation due to collisional cooling does not seem to be significant, at least in this case. Only with the addition of a thin layer of oil did copper powder show clustering similar to that seen with the glass beads. Even more interesting, clustering was prevented by the addition of nanoscale asperities to the glass beads.

Royer [31] found that surface roughness may be a leading indicator for these cohesive forces. Surfaces, where roughness was significant, such as for the copper powder and the glass beads with nanoscale asperities, seem to prevent particle clustering. Particles with smooth surfaces such as that of the glass beads and the silver-coated glass beads were prone to particle clustering. Large-scale roughness may reduce sliding, leading to different rotational collisional dynamics. In other words, surface roughness does not lower the granular temperature as well as smooth surfaces. When the granular temperature is low enough, cohesive forces such as van der Waals, electrostatics or capillary-like forces can take over.

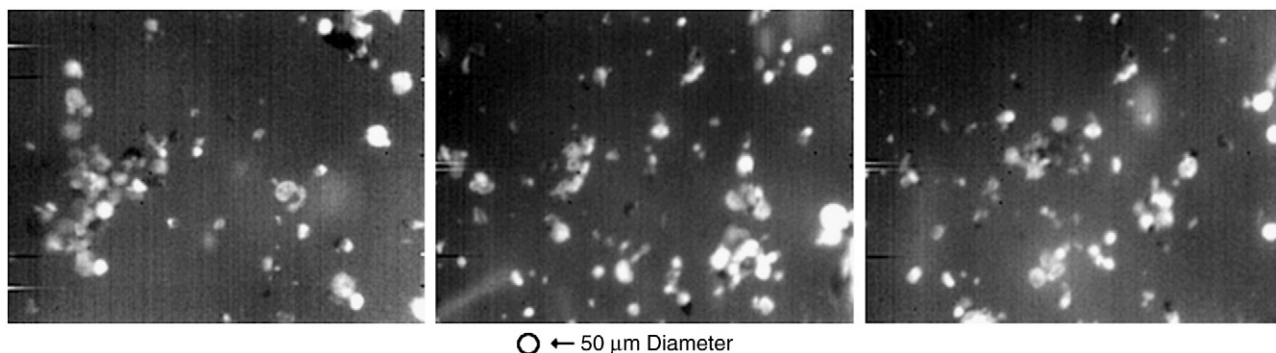


Fig. 13. Several consecutive frames of FCC catalyst in the bed region of a 6-in. (16-cm) diameter fluidized bed at a superficial gas velocity of 2 ft/s (0.61 m/s). Images were collected at 4000 fps with a 20 μ s exposure time. 5 $\times$ magnification compared to Fig. 12.

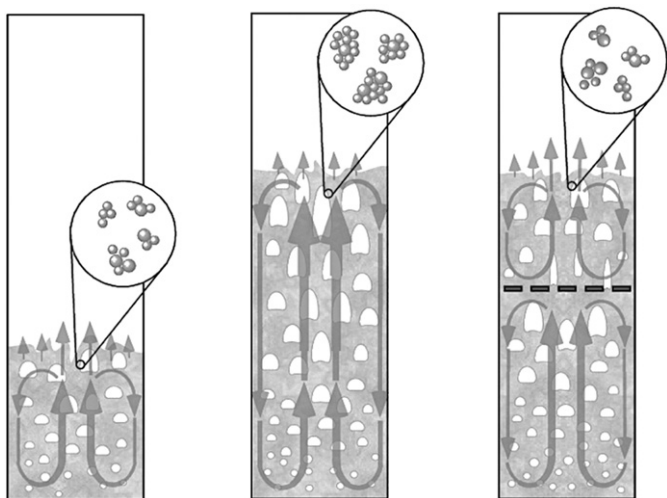


Fig. 14. Conception of a proposed cluster formation model for a shallow bed (left), tall bed (middle) and tall bed with baffle (right).

Both Hopkins et. al. [34,35] and Rice, et al. [36] have conducted discrete element simulations that showed some shear was needed to have particles form particle clusters or particle clouds. Hopkins found that cluster size was dependent on the coefficient of restitution. As the particles became more elastic, larger particle clusters or clouds were observed. However, Hopkins' model did not have any cohesive forces. Rice's model, which had a van der Waals cohesive force, showed similar results, but found that particle clusters were more prevalent in systems that are more polydispersed. Both models suggest that a short-range reach of cohesive forces may cause particle clustering. Particle shear with some form of collisional dissipation may be needed to get the particles "preconditioned" for clustering.

Based on the observations with polyethylene and FCC powders in fluidized beds along with Royer's powder drop experiments, the formation of particle clusters appears to be a multiple-step process, as shown in Fig. 15. Some shear is needed to promote collisional cooling or dissipation. In a fluidized bed, this would be near or at the gas jets of a grid plate or sparger or the bubbles themselves. Indeed, the only evidence of particle clusters in fluidized beds offered here are the clusters located near the bubbles.

If enough energy is dissipated in the collision and not redirected into rotation, the granular temperature (and rotational energy) may decrease enough such that various cohesive forces dominate the hydrodynamics. Those forces could be electrostatic, capillary or van der Waals. For polyethylene, one would suspect electrostatics to be significant. In fact, one could argue that collisional cooling may not be a requirement for electrostatic induced clustering. For FCC catalyst, it could be capillary-like forces with a water monolayer (not liquid

bridging in the strictest sense). For even smaller particles, van der Waals may have a significant role.

As noted above, particle cluster size could be a function of collisional cooling or dissipation. For polyethylene, the surfaces are smooth and the coefficient of restitution is low. Most of the collisions probably resulted in a direct decrease in the granular temperature, which resulted in more particles being in proximity to each other where dominant cohesive forces persist – presumably electrostatics. FCC catalyst powder is rougher and has a higher coefficient of restitution. Collisions may lead to more of a redirection of rotation and translation spatial components. The granular temperature is less affected and smaller particle clusters develop.

Other studies [37,38] have suggested that dissipation due to drag could reduce the granular temperature of particles. This could certainly be the case in a riser where drag has a significant role in the overall hydrodynamics. In some cases, particle clusters begin to affect gas-phase hydrodynamics and vice versa [37]. Streamers or macro-clusters result due to the drag force on the clusters coupling with the gas-phase hydrodynamics. In the case of risers, these streams can be significant.

However, Royer [31] showed that clustering can readily occur without drag. Thus, no one mechanism appears to be applicable to all cases. It is more likely that particle clustering is a function of a multitude of mechanisms which are dependent on the particle properties and the environment. In fluidized beds, collisional cooling and cohesive forces may play a key role as with the powder drop experiments of Royer. In risers, the role of drag may be key in, at least, the precursor for the formation of these clusters.

Thus, the formation of micro-clusters may be the result of sequences of events. In fluidized beds, it may start with collisional cooling or dissipation to clustering resulting from cohesive forces. In risers, dissipation due to drag could lead to a lowering of granular temperature such that the environment may more resemble a fluidized bed where collisional cooling and cohesive forces start playing a role. These macro-clusters may still form without the presences of micro-clusters, but the presence of micro-clusters may make the macro-clusters more stable and possibly more significant.

6. Conclusions

Using a high-speed video camera with a modified rigid boroscope allowed particle clusters to be visualized, beyond the column wall, in and above a fluidized bed of polyethylene and FCC catalyst powders. The fact that clusters were observed in a fluidized bed suggests that hydrodynamics may not be solely responsible for cluster formation. Cohesive forces such as electrostatics, capillary and van der Waals forces, may play a significant role in particle cluster formation. The proposed mechanism suggests that particle shear results in collisional cooling that allows the granular temperature to decay to a point where these cohesive forces can dominate. In some cases, dissipation from drag may be needed to start this process. The decrease in the granular temperature appears to be dependent on the particle properties and surface morphology. Collisions that only redirect the particle or increase particle rotation limits this reduction in the granular temperature such that cohesive forces have less of an impact. In the case of risers where large shear streams are prevalent, these clusters and the corresponding drag forces may result in the formation of large streamers.

Acknowledgements

The authors would like to thank John Roper, Scott Waitukaitis, Sid Nagel, and Heinrich Jaeger of the University of Chicago for their invaluable discussion on particle clustering. The authors would like to thank Alexander Mychkovsky and Steve Ceccio of University of Michigan for their help with consultation with the development of the boroscope. The authors also thank the National Energy Technology

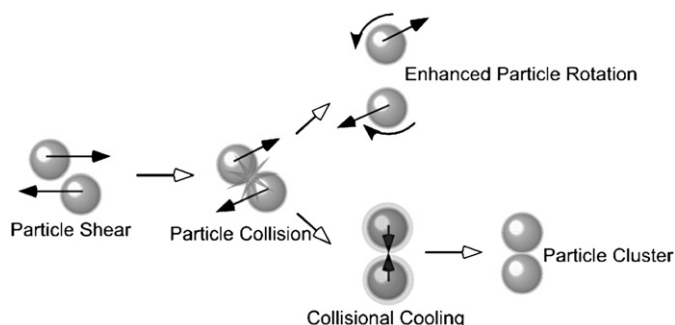


Fig. 15. Conception of the formation of particle clusters due to granular temperature relaxation via collisional cooling and subsequent cohesive interaction.

Lab and the Department of Energy for their funding under DE-FC26-07NT43098 as well as their expertise.

References

- [1] R.H. Wilhelm, M. Kwauk, Fluidization of solid particles, *Chemical Engineering Progress* 44 (1948) 201–218.
- [2] J. Happel, R. Pfeffer, The motion of two spheres following each other in a viscous fluid, *AIChE Journal* 6 (1960) 129–133.
- [3] K.O.L.F. Jayaweera, B.J. Mason, G.W. Slack, The behavior of clusters of spheres falling in a viscous fluid, *Journal of Fluid Mechanics* 20 (1964) 121–128.
- [4] R. Johne, Einfluß der Konzentration einer monodispersen Suspension auf die Sinkgeschwindigkeit ihrer Teilchen, *Chemie Ingenieur Technik* 38 (1966) 428–430.
- [5] J.M. Crowley, Clumping instability of a falling horizontal lattice, *Physics of Fluids* 19 (1976) 1296–1301.
- [6] A.L. Graham, R.B. Bird, Particle clusters in concentrated suspensions. 1. Experimental observations of particle clusters, *Industrial and Engineering Chemistry Fundamentals* 23 (1984) 406–410.
- [7] A.F. Fortes, D.D. Joseph, T.S. Lundgren, Nonlinear mechanics of fluidization of beds of spherical particles, *Journal of Fluid Mechanics* 177 (1987) 467–483.
- [8] M.Kwauk Wang, H. Li, Fluidization of fine particles, *Chemical Engineering Science* 53 (1998) 377–395.
- [9] U.-M. Chen, C.-S. Jang, P. Cai, L.-S. Fan, On the formation and disintegration of particle clusters in a liquid–solid transport bed, *Chemical Engineering Science* 46 (1991) 2253–2268.
- [10] B.M. Kaye, R.P. Boardman, Cluster formation in dilute suspensions, in *Proc. Symp. on the Interaction between Fluids and Particles*, Institution of Chemical Engineers London (1962) 17–21.
- [11] J. Yerushalmi, D.H. Tuner, A.M. Squires, The fast fluidized bed, *Industrial & Engineering Chemistry Process Design and Development* 15 (1976) 47–53.
- [12] J. Yerushalmi, N.J. Cankurt, Further studies of the regimes of fluidization, *Powder Technology* 24 (1979) 187–205.
- [13] Matsen, Mechanisms of choking and entrainment, *Powder Technology* 32 (1982) 21–33.
- [14] D. Geldart, A.C.Y. Wong, Entrainment of particles from fluidized beds of fine powder, *AIChE Symposium Series* 255 (1987) 1.
- [15] J. Baeyens, D. Geldart, S.Y. Wu, Elutriation of fines from gas fluidized beds of Geldart A-type powders – effect of adding superfines, *Powder technology* 71 (1992) 71–80.
- [16] J.-H. Choi, J.-M. Suh, I.-Y. Chang, D.-W. Shun, C.-K. Yi, J.-E. Son, S.-D. Kim, The effect of fine particles on elutriation of coarse particles in a gas fluidized bed, *Powder technology* 121 (2002) 190–194.
- [17] J. Li, T. Nakazato, K. Kato, Effect of cohesive powders on the elutriation of particles from a fluid bed, *Chemical Engineering Science* 59 (2004) 2777–2782.
- [18] T.J. O'Brien, M. Syamlal, Particle cluster effects in the numerical simulation of a circulating fluidized bed, in: A.A. Avidan (Ed.), *Preprint Volume for Circulating Fluidized Beds IV*, AIChE, Somerset, 1993, pp. 345–350.
- [19] R. Bader, J. Findlay, T.M. Knowlton, *Circulating Fluidized Bed Technology II*, in: P. Basu, J. Large (Eds.), Pergamon Press, Oxford, UK, 1988, pp. 123–137.
- [20] Agrawal, et al., The role of meso-scale structures in rapid gas–solid flows, *Journal of Fluid Mechanics* 445 (2001) 151–185.
- [21] Guenther, Breault, Wavelet analysis to characterize cluster dynamics in a circulating fluidized bed, *Powder technology* 173 (3) (2007) 163–173.
- [22] Horio, Kuroki, 3-Dimensional flow visualization of dilutely dispersed solids in bubbling and circulating fluidized-beds, *Chemical Engineering Science* 49 (15) (1994) 2413–2421.
- [23] Noymer, Glicksman, Cluster motion and particle-convective heat transfer at the wall of a circulating fluidized bed, *International Journal of Heat and Mass Transfer* 41 (1) (1998) 147–158.
- [24] Rhodes, et al., Particle motion at the wall of a circulating fluidized bed, *Powder Technology* 70 (3) (1992) 207–214.
- [25] A.K. Sharma, J.M. Matsen, K. Tuzla, J.C. Chen, A correlation for solid fraction in clusters in fast-fluidized beds, in: M. Kwauk, J. Li, W.-C. Yang (Eds.), *Fluidization X*, 2001, p. 301, Beijing, China.
- [26] R. Hays, S.B.R. Karri, R. Cocco, T.M. Knowlton, Small particles cluster formation in fluidized beds and its effect on entrainment, *Circulating Fluidized Bed* 9 (2008) 1–5.
- [27] Subbarao, Clusters and lean-phase behaviour, *Powder Technology* (2–3) (1986) 101–107.
- [28] J. Visser, An invited review – van der Waals and other cohesive forces affecting powder fluidization, *Powder Technology* 58 (1989) 1.
- [29] Horio and Clift, A note on terminology: “clusters” and “agglomerates”, *Powder Technology* 70 (1992) 195–196.
- [30] K.D. Squires, J.K. Eaton, Particle responses to turbulence modification in isotropic turbulence, *Physics of Fluids A-Fluid Dynamics* 2 (1991) 1191.
- [31] Wylie, Koch, Particle clustering due to hydrodynamic interactions, *Physics of Fluids* 12 (2000) 964.
- [32] J.R. Royer, D.J. Evans, L. Oyarte, Q. Guo, E. Kapit, M.E. Möbius, S.R. Waitukaitis, H.M. Jaeger, High-speed tracking of rupture and clustering in freely falling granular streams, *Nature* 459 (2009) 1110–1113.
- [33] G. Kuwabara, K. Kono, Restitution coefficient in a collision between two spheres, *Journal of Applied Physics* 26 (1987) 1230.
- [34] M.A. Hopkins, M.Y. Louge, Inelastic microstructure in rapid granular flows of smooth disks, *Physics of Fluids* 3 (1991) 47–57.
- [35] M.A. Hopkins, J.T. Jenkins, M.Y. Louge, On the structure of three-dimensional shear flows, *Mechanics of Materials* 16 (1993) 179–187.
- [36] Rice, B.R., Hrenya, C.M., “Clustering in rapid granular flows of binary and continuous particle size distributions”, (2009) in press.
- [37] E. Helland, H. Bournot, R. Ocellli, L. Tadriss, Drag reduction and cluster formation in a circulating fluidised bed, *Chemical Engineering Science* 62 (2007) 148–158.
- [38] K. Agrawal, P.N. Loezos, M. Syamlal, S. Sundaresan, The role of meso-scale structures in rapid gas–solid flows, *Journal of Fluid Mechanics* 445 (2001) 151–185.