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Radial Distribution of Particle Clusters in Down Flow Reactors

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RADIAL DISTRIBUTION OF PARTICLE CLUSTERS IN DOWN FLOW REACTORS

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ABSTRACT

Particle clustering is of major importance in down flow reactors having a profound influence on some fundamental properties of the flowing suspension such as particle slip velocity. The goal of this study is to provide further evidence to support the formation of clusters using the novel CREC-GS-Optiprobes. This sensor is equipped with GRIN lenses and introduces minimum intrusion effects. Results reported include radial distributions of particle cluster size and velocity under various gas flow superficial velocities and suspension densities. Micro-scale and macro-scale flow structures are advanced on the basis of the reported data.

INTRODUCTION

Particle clustering is a dominant feature of upward fast fluidization in riser reactors (Nova et *al.* (1), Jin et *al.* (2)). In downflow reactors, however, the possible aggregation of particles is still an open question and one that is necessary to clear up. In downflow units, the solid particle flow is assisted by gravity, given that particle flow and gravity have a coincident direction.

In addition, the solid particles experience a so-called "slip velocity" because as they move in the downer, they evolve at a higher velocity than the gas phase. As predicted by the law of particle mechanics, the slip velocity for single particles can be calculated as a balance of forces exerted on a particle. However, in downers where there is a population of particles moving simultaneously, particles may influence each other while falling side by side or while travelling one behind the other. Under these conditions, there may be a drag force reduction with mass increment due to the consolidation of agglomerates and all this may contribute to a rise in slip velocity. In respect to this issue, Happel and Brenner (3) observed an increase in velocity while two particles were falling side by side or one particle was moving behind the other. Zheng *et al*. (4), claimed that if two identical spheres fall one behind the other with a separation distance smaller than a critical distance, their velocity may be increased by up to 50%.There may also be other contributing factors. For instance, if the surface of the particle is rough, the wake separation is delayed with a consequently smaller drag, allowing the particles to evolve at a faster velocity than the expected terminal velocity value. In addition, surface roughness causes the attached boundary layer to easily change from laminar to turbulent. As a result, at low Reynolds numbers, the drag force is reduced with a subsequent increase in velocity (Clift, *et al.*, (5)). In regards to particle agglomeration in down-flow reactors, there is the suggestion that agglomeration is a phenomenon potentially occurring in those units (Bai et al. (6), Zhu and Wei (7)). There is, however, the need to elucidate with direct experimental evidence, the existence of these agglomerates. Sobocinski *et al*. (8), and Krol *et al*. (9) advanced the view that these agglomerates are in fact strings of particles. It is the goal of the present study to provide further evidence about the formation of these clusters in down-flow units.

To accomplish this, a new CREC-GS-Optiprobe has been developed, with the focus on the advanced design and calibration of this device (Nova et *al.* (10)). In this work, the CREC-GS-Optiprobes are applied for the characterization of cluster phenomena in downer reactors and its variation with radial position.

EXPERIMENTAL METHODOLOGY

One of the main objectives of the present study was to obtain fluid dynamic information on the particle clustering phenomena in a downflow unit using a CREC-GS-Optiprobe. The study comprised the estimation of the slip velocity from cross-correlation analysis, the calculation of solid hold-up with a simplified model and the estimation of the cluster size through peak width analysis. For estimation of the cluster velocity, the cross-correlation function between the data of two time series was used. The time series came from two CREC-GS-Optiprobes axially spaced along the flow stream in the downer section. The time delay, corresponding to the maximum value of the cross-correlation function, gave the particle transit time between the two sensors. The length of the distance between the measuring regions of the upper and lower sensors and the particle transit time between both sensors were used to estimate the velocity of the cluster. The superficial gas velocity in the downer section was calculated from the gas flow rate, measured at the outlet of the downer section of the experimental set-up, and the cross-sectional area of the column. For the next part of the calculations, the solid hold-up and the gas void fraction were required. They were estimated with a simplified model that involved the inclusion of the average particle velocity. Next, the slip velocity was calculated as the difference between the velocity of the particles and the velocity of the superficial gas velocity divided by the gas void fraction. The evaluation of the cluster size was based on measurements of the widths of the signal peaks. All the signal peaks were considered in the peak width analysis if they complied with the constraint of being at least as tall as 95% of the highest peak in the signal. Signal peak heights were considered as the distance between baselines, set at zero volts, and the peak's maximums. The peak widths were measured at a horizontal reference level, crossing the signal at 50% of the height of the highest peak in the signal. The width of an individual peak was obtained as a time interval in seconds corresponding to the time that the cluster took to cross the length of the diameter of the focal point of the probe (probe's measuring region). The cluster size was then evaluated from the time interval, representing the width of the cluster, the velocity of the cluster calculated from the cross-correlation analysis, the length of the diameter of the measuring region of the probe and the average diameter of the solid particles. The cluster size was estimated in both time series *X* and *Y* from the upper and the lower probes respectively.

The operation of the experimental unit required that the optoelectronic system, including the CREC-GS-Optiprobe, perform the data acquisition. At the same time, the data acquisition system needed certain software programs in order to function. Additionally, due to the very large amounts of data generated, these programs were essential for analysis of the data. Three LabVIEW 6.1 programs were implemented for this purpose: the program "Probe Signal Sampling.vi" for data acquisition in the experimental unit, another program called "Cross Correlation Function Hi-Speed.vi" for estimation of the cluster velocity through calculation of the average transit time from the cross-correlation function, and the last one called "Pulse Width.vi" for estimation of the cluster size by measuring the widths of the peaks of the signals. Additional information regarding data acquisition programs is reported in Nova (11).

Experimental Runs

The operating conditions of the riser-downer unit were set according to a combination of independent variable values such as gas volumetric flow rate and solid mass flow rate. The experimental unit employed to simulate the hydrodynamics of a downflow reactor is described in Nova (10). In this unit, the gas volumetric flow rate can be controlled with various pressure regulators and valves to obtain the desired value with the help of a rotameter flowmeter connected at the gas outlet of the downer section. The solid mass flow rate can be adjusted through a ball valve feeding the particles into the riser section. The ball valve can be positioned at any angle between 0 and 90 degrees to regulate the amount of solids entering the system. The temperature of the compressed air varied from 19 to 25 ºC. The absolute operating pressure of the downer section varied from 103 to 112 kPa. The manometric operating pressure was measured just below the downer air injector. FCC particles with a mean particle diameter of 76 μm were employed as the solid phase while air was used as the fluidizing medium. Regarding the experiments developed in the present study, the independent variables investigated were the superficial gas velocity and the solids mass flow rate. Table 1 summarizes the conditions of the independent variables considered for these experiments.

Table1 Ranges of independent variables investigated.

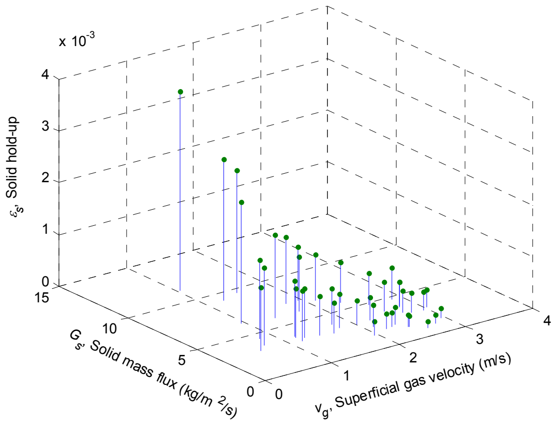
Independent Variable	Minimum Value	Maximum Value
v_g, Superficial Gas Velocity (m/s)	0.7	5
G_s, Solid Mass Flux (kg/m^2/s)	0.9	103

Experimental Results and Analysis

A total of 105 experimental runs were effected in the downer section of the riser-downer unit. The operating conditions at each particular experimental run were set according to a combination of gas flow and solid particle flux. The 105 experimental runs can be divided into three main groups. An initial group of 42 experimental runs were completed with readings taken exclusively at the central axis of the downflow section. The main goal of this first group of 42 runs was to give the physical evidence to support the theory of cluster formation in downflow reactors formulated by Krol et al (9) as well as validate the capability of the CREC-GS-Optiprobe design to detect and quantify the cluster size and the cluster velocity in downers under a wide range of operating conditions. This group of 42 experimental runs is characterized for its operation at low solid concentrations ranging from solid hold-ups (ε_s) of 0.0001 to 0.0038. Once the design was validated, a second group of 23 runs was completed along the radius of the downflow fluidized bed unit to study the radial particle velocity profiles and the radial cluster size distribution. This group of 23 experimental runs also permitted the study of the effects of the various operating parameters of the riser-downer unit on the radial cluster velocity profiles and on the radial particle size distribution. This group of experimental runs also permitted the optimization of the design and operation of the riser-downer unit as well as the implementation and validation of the various software programs used for data acquisition and analysis. Medium solid concentrations ranging from solid hold-ups (ε_s) of 0.0002 to 0.0077 are characteristic of this group. After the operating parameters on the radial cluster velocity profiles and on the radial particle size distribution were well understood and a robust system of software programs was implemented, a group of 40 experimental runs were completed to study the relationship between the cluster size and the slip velocity in a downflow unit under a wide range of operating conditions. This group is characterized as using higher solid concentrations ranging from solid hold-ups (ε_s) of 0.0018 to 0.0240.

Cluster Average Properties

The results of the at group of 42 experimental runs (Figure 1) with GS-Optiprobes placed in the centre of the down flow unit allowed the calculation of both the cluster slip velocity and the solid hold-up, ε_s, defined as a fraction of the total volume in the pipe occupied by the solid phase. The slip velocities for the 42 experimental runs were estimated as a function of the size of the cluster are reported in Figure 2. This figure shows that the slip velocity of the clusters is a function of the size of the cluster: then the larger the cluster mass the greater the expected slip velocity. In general, particle agglomerates containing between 2.0 and 6.3 particles were estimated for down-flow systems with FCC catalyst ($d_{p,av}$ = 76 µm). Under the operating conditions covered particles should evolve in the downer as strings, clusters or agglomerates with a relatively large distribution of cluster length, ranging from 150 µm to 480 µm, and with most of these strings having from 2.9 to 5.5 times the average particle diameter in length. Furthermore, as shown in Figure 2, the slip velocity (u_{slip}) follows an almost linear relationship with respect to the number of particles conforming the cluster (N), as is expected according to the theoretical considerations of Krol, *et al*. (9).



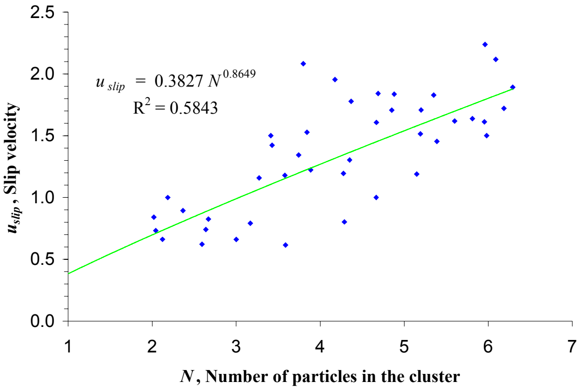


Figure 1 3-D representation of the operating conditions for the forty-two experimental runs: solid hold-up versus solid mass flux and superficial gas velocity.

Figure 2 Cluster slip velocity versus the number of particles contained in the cluster.

Radial Velocity Profiles and the Radial Cluster Size Distribution

For the study of the radial particle velocity profiles and the radial size distribution function a group of 23 runs in the downer unit was effected. The operating conditions of these three experimental runs are given in Table 2, with a superficial gas velocity being essentially the same. Results obtained using the CREC-GS-

Optiprobes are reported in plots in terms of the cluster velocity, slip velocity and cluster size over the column radius. The load of solids fed to the downer system was increased from one run to another run to see its effect on the solid hold-up and on various radial profiles (e.g., cluster velocity, slip velocity, cluster size). Figures 3 and 4 show the results of typical cluster velocity profiles, the slip velocity profile over the column radius for the first run fro Runs 1 and 2 respectively.

Table.2 Ranges of independent variables investigated in Runs 1, 2 and 3

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	Run 1	Run 2
u_p, Superficial Gas Velocity (m/s)	1.8	1.6
G_s, Solid Mass Flux (kg/m^2/s)	8.2	63.4
Pressure (kPa)	106.5	107.5
Temperature (ºC)	21	21
ε_s, Solid hold-up (-)	0.0010	0.0077

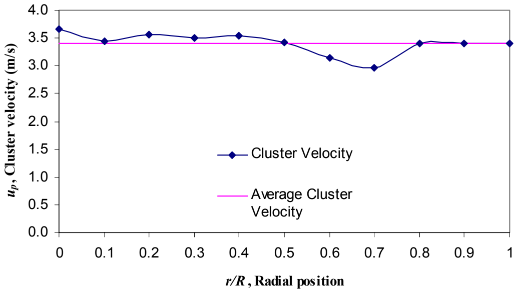


Figure 3 Radial cluster velocity profile for Run 1 (v_g = 1.8 m/s, Gs = 8.2 kg/(m^2-s)).

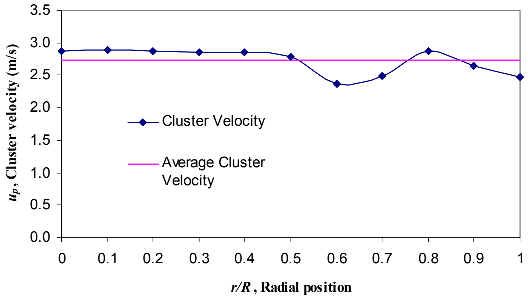
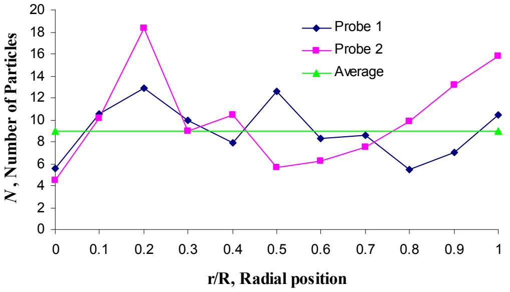


Figure 4 Radial slip velocity profile for Run 2 (v_g = 1.6 m/s, Gs = 63.4 kg/(m^2-s)).

It can be observed from Figures 3 and 4 that practically uniform cluster velocity profiles over the column radius are obtained. These profiles suggests practically a plug flow solid patterns over the entire cross section of the column. No significant effect at the close-to-wall regions is observed and this was considered a consequence of the relatively small column diameter used in this study. These results are consistent with the plug flow profiles predictions from CFD simulations for a downer section using two CREC-GS-Optiprobes (Nova, (12)) and also to some degree in agreement with results reported in Zhu, *et al*., (13) with small differences at the centre of the downer and in the close-to-wall region. In Zhu, *et al*. (13), the particle velocities are slightly lower at the centre of the downer, somewhat higher at the close-to-wall region and smaller at the near wall region.



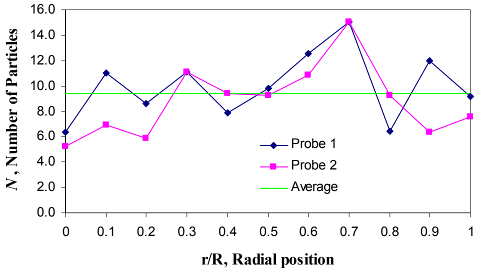


Figure 5 Radial cluster size distribution for

Figure 6 **Radial** cluster size distribution for

Run 1 (v_g = 1.8 m/s, Gs = 8.2 kg/(m^2-s)). Run 2 (v_g = 1.6 m/s, Gs = 63.4 kg/(m^2-s)).

The results of flock and cluster size from the three groups of experimental runs are reported in Figure 7. The general observed trend of the slip velocity is to increase as the clusters grow from 2 to 6 particles. After a gradual grow, the slip velocity remains constant for flock sizes ranging from 7 to 20. On the basis of this data a transition zone is found where the concentration of clusters increases to a level where they start creating assembles or flocks of clusters. This transition zone is located between the 6 to 7 particle range.

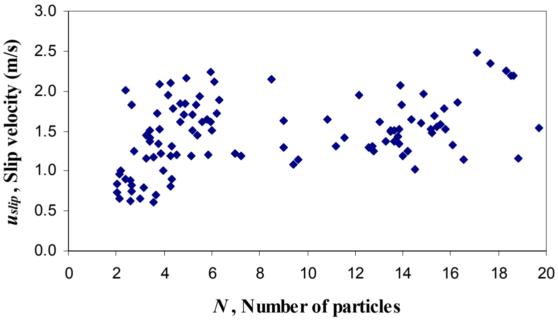


Figure 7. Cluster slip velocity versus the number of particles contained in the versus solid mass flux and superficial gas velocity experimental runs.

These results suggest that at higher solid concentrations a characteristic particle flow pattern exists where the individual clusters of particles (leading particle and trailing particles) travel in "flocks" (i.e. cluster ensemble). Each individual "flock" of particle clusters remains confined to well defined limits, apart from other flocks, interacting only when they merge together to form a greater flock. In this proposed gas-solid structure, forces in equilibrium between the individual clusters, keep the flocks (cluster ensemble) loosely together allowing the permeation of gas between them without creating an increase in the overall drag. Thus, in the downflow unit, a complex mechanism takes place inside the limits of each of the flocks of clusters, similar to the one proposed by Krol et al (9), Krol and de Lasa (14). Inside the "flock" (cluster ensemble), the clusters are strings formed from single particles due to inter-particle and particle-fluid dynamic forces. The string configuration of the cluster inside the flock is the result of the tendency of the particles to travel in the wakes of other particles experiencing, in this way, a smaller resistance due to a drag reduction. Inside these pockets of high particle cluster populations, the clusters grow and collapse as a result of a dynamic equilibrium process between them. In this way, with this new proposed structure, it is possible to justify the experimental results where there is simultaneous coexistence of wide signal peaks, coming from larger particle structures (flocks of clusters with equivalent lengths ranging from 10 to 20 particle diameters, Figure 7), with the detection of low slip velocities, belonging to short clusters (strings of particles with 2 to 6 particles, Figure 2). It is worth mentioning that the downflow structure proposed in this study is the result of observations and measurements in a downflow unit containing dry FCC particles with an average diameter of 76 μm.

Thus, the reported data in Figures 2 and 7 for the fully developed region of a downflow fluidized bed (15) can be explained as described in Figure 8 as follows:

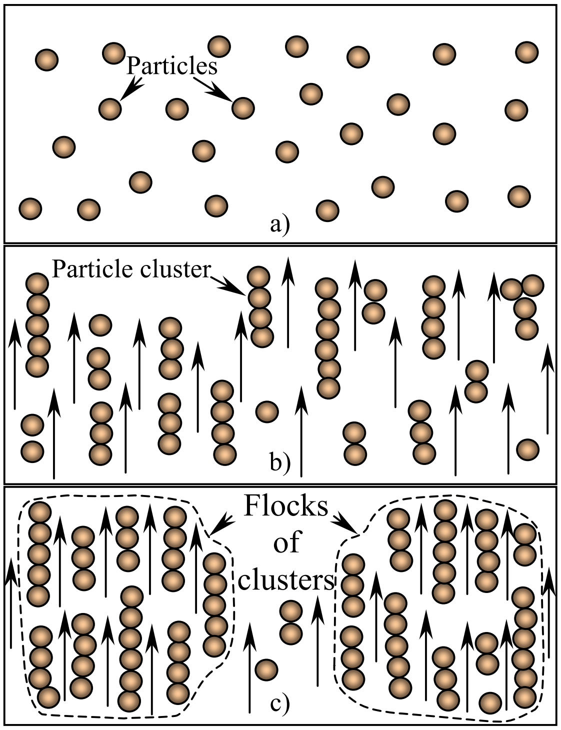


Figure 8. Schematic representation of cluster formation and cluster densification in a constant velocity section of a downer: a) dilute downwards flow pattern, b) downwards cluster flow pattern, and c) downwards flock flow pattern. The upward vectors represent relative gas velocity between the gas and the solid particles.

a) *Dilute downwards flow pattern (G_s < 8 kg/(m^2.s))*: At very low solid particle concentrations, the flow pattern preferred in downflow units is the one comprised of single dispersed particles traveling downwards. The particle slip velocity can be predicted from simple laws of particle mechanics.

b) Downwards cluster flow pattern(8 kg/(m^2.s) < G_s < 24 kg/(m^2.s)): As the load of particles continues increasing in a downward fluidized bed, the solid particle concentration gets augmented and there is a higher probability of particle-particle interaction with this leading to strings of two or three particles. If the load of particles continues increasing, the strings of particles grow and dominate the flow structure, forming particle clusters of four to six particles. During this downward flow-regime, the slip velocity can no longer be predicted from the laws of particle mechanics for single particles and; more complex models involving the string of particles are required (9).

c) Downwards flock flow pattern (G_s> 24 kg/(m^2.s)): As the particle concentration continues increasing, the clusters start interacting more frequently at a point where they cannot grow indefinitely, and in order to further reduce the drag resistance, they experience a clusters densification by getting closer and forming flocks of clusters.

CONCLUSIONS

a) A CREC-GS-Optiprobe, of the reflective type with an incorporated GRIN lens, was successfully implemented to characterize the gas-solid flow patterns in a down flow unit. This sensor is able to perform measurements of particle cluster velocity in a region far away from the tip of the probe, resulting in minimum distortion of the flow patterns and, as a consequence, negligible intrusion effects.

b) Data of the present study show that, at low particle concentrations, these strings of particles follow a slip velocity-cluster length relationship, consistent with that expected to apply from basic fluid dynamic principles for this type of agglomerate.

c) Results from medium solid particles concentrations suggest the existence of more complex structures than simple clusters since large particle sizes are obtained simultaneously with low particle slip velocities. A structure comprised of flocks of clusters explains this phenomenon.

d) Data from cluster (or flock) velocity profile and cluster slip velocity studies revealed the existence of practically quite uniform velocities profiles along the radius of the downer with the cluster size also remains practically constant along the radius of the downer.

ACKNOWLEDGEMENTS

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NOTATION

G_s, solid flux , kg/(m^2.s)
$d_{pav,}$ average particle sixe (microns)
N, number of particles in a cluster (-)
R, radial position in the downer (m)
R, radius of the downer (m)
u_p, particle velocity, m/s
u_s, particle slip velocity , m/s
Vg, gas superficial velocity , m/s
ε_s, solid particle concentration (-)

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