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FLUIDIZATION BEHAVIOR IN A GAS-SOLID FLUIDIZED BED
WITH THERMALLY INDUCED INTER-PARTICLE FORCES

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ABSTRACT

In this work, a new approach for increasing and controlling inter-particle forces (IPFs) was applied. This method used a spherical inert particle coated with a polymer material having a low glass transition temperature. Since IPFs depend on the temperature of the coated particles, they can be easily controlled by the temperature of the inlet air. For this reason, the temperature of the system was varied uniformly near the glass transition temperature of the polymer, between 20 – 40°C, to investigate the effect of IPFs on fluidization behavior at low and high gas velocities.

INTRODUCTION

Particle size, shape, roughness and density as well as inter-particle forces (IPFs) are among the most important parameters affecting the flow dynamics of powder materials. In regard to the significance of IPFs, for instance, there is no doubt that IPFs dominate the fluidization behavior of Geldart Group C particles, which leads to a completely different behavior compared to the other groups of Geldart’s classification with low or no IPFs.

Investigations into the influence of IPFs on the behavior of a gas-solid fluidized bed have been carried out using different methods. Most importantly, however, controlling the level of IPFs to have a uniform cohesion throughout the particulate media is not an easy task. Methods that have been used include the following: increasing the amount of Van der Waals forces by decreasing the mean size of particles (1); increasing the amount of capillary forces by the addition of a cohesive agent into the bed (2); creation of a magnetic field around the bed (3) or raising the bed temperature to a high value (4).

Each of these methods has specific difficulties in practice. For the first method it is very difficult to control particle shape and surface roughness. Due to these difficulties, the use of Van der Waals forces to study the effect of IPFs on fluidization characteristics is a complex procedure. By increasing the size of particles, the hydrodynamic forces become dominant compared to the IPFs. For larger particles, an increase in IPFs must be induced by the addition of a cohesive agent into the particulate system. One of the most popular methods to conduct this technique is by the presence of wet capillary bonds created by an interstitial liquid between the particles. The problem with this approach is that it is challenging to have a uniform distribution of the agent throughout the whole bed, which leads to force anisotropy
inside the particulate system (5). The other problem with this approach is that it restricts the fluidization study at low superficial gas velocities. To employ the third method an expensive set-up is needed to generate a magnetic field with the help of a costly electromagnetic coil system. The other problem of this approach is that the ferromagnetic particles attract themselves when they are parallel to the magnetic field and repel each other when they are perpendicular, which, consequently, causes anisotropic attraction/repulsion in the bulk materials (6). For the last method, the first problem is that it is costly to have an apparatus operating at high temperatures. Secondly, the lack of proper measurement techniques at high temperatures is the other difficulty with this strategy (7).

In this work, a novel approach is proposed to induce IPFs inside a particulate media. It involves large particles that are not significantly influenced by the Van der Walls forces and other colloidal interactions. This technique uses a copolymer of PMMA/PEA (Poly Methyl MethAcrylate/Poly Ethyl Acrylate) contained in a polymer suspension called Eudragit NE30D. The copolymer, which is characterized by a low glass transition temperature, around 9°C, is coated on inert particles by an atomization process. By changing the ambient temperature to which the coated particles are exposed, the polymer adhesion/friction parameters and Young Modulus are modified in a way that the observed cohesive IPFs between the particles are changed significantly. Accordingly, the cohesion between particles can be adjusted by temperature in a stable and reproducible manner. The advantage of this method is that it does not necessitate the addition of any liquid phase, which has to be uniformly distributed into the particulate system. Besides, it allows the fluidization study to be carried out at both low and high superficial gas velocities. In contrast to the magnetic interactions, the forces are not dominant in only one direction, but are located at each contact between the particles. Thus, there is no force anisotropy present except if the temperature is not uniform in the system. The last priority of this technique is that it can be conducted at low temperatures and IPFs are changed in a completely controlled manner by merely a small increment in the system temperature.

METHODOLOGY

Particle Coating Process

The experimental work necessitates having inert particles as base particles, which can accept the copolymer of PMMA/PEA as the coating. A 450-720 μm cut of spherical sugar beads \((d_p=580\mu m, \rho_p=1575 \text{ kg/m}^3)\), which lie in Geldart Group B particles, were chosen as inert particles in this work.

Sugar beads were coated by an atomization process with a polymer suspension in water and dried simultaneously to obtain a uniform coating on the particle surface. It was achieved in a spheronomizer, which allowed the introduction of air under the rotating disc located inside the bowl. Table 1 presents operating parameters associated with the coating process of the particles. Heated air, which was passed through an electrical heater before entering the processing chamber, was used for adjusting the temperature to the desired operating setpoint and also for concurrent drying of the particles. The temperature was controlled with the help of a controller coupled with an infrared cell, which measured the surface temperature of the particles. In addition, a thermocouple located under the spheronomizer disc allowed for measuring the air entrance temperature. The air flow rate was adjusted to the
desired value with the help of a rotameter. The coating solution (Water 0.21 kg; PMMA/PEA 0.086 kg; Nonoxynol100 0.004 kg) was added by atomization onto the particles with a Schlick 970 series two-substance atomizer. The atomizer was fed with the solution by a peristaltic pump with a flow rate approximately equal to 1 g/min and compressed air, which allowed the formation of fine droplets. The atomizer gun was arranged in such a manner that the tip of the nozzle was placed at approximately 4 cm from the torus surface to avoid coating losses on the parts of the equipment. The characteristics of the final product are summarized in Table 2.

<table>
<thead>
<tr>
<th>Table 1. Spheronizer’s Operating Parameters</th>
<th>Table 2. Final Particles Coating Characteristics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Disc rotational rate (rpm)</td>
<td>230</td>
</tr>
<tr>
<td>Air flow rate (cfm)</td>
<td>25</td>
</tr>
<tr>
<td>Air temperature (°C)</td>
<td>30</td>
</tr>
<tr>
<td>Solution flow rate (g/min)</td>
<td>1</td>
</tr>
<tr>
<td>Atomization pressure (bar)</td>
<td>2</td>
</tr>
</tbody>
</table>

**Experimental Set-up and Procedure for Fluidization Study**

The experimental set-up used for the fluidization study consisted of a fluidization column, which was constructed with a transparent Plexiglas tube with 0.152 m I.D. and 1.5 m in height. Dried and filtered air was introduced into the bed through a perforated plate as the distributor. It contained holes 1 mm in diameter arranged in a triangular pitch.

Air was heated with the help of an electrical heater before entering the fluidizing column. Accordingly, it was used to adjust the temperature of the bed to a desired value. Temperature was controlled by means of a PI controller driven by a thermocouple constantly immersed in the bed. A thermocouple located at the windbox allowed measuring the air entrance temperature. Furthermore, the air flow rate was controlled with a calibrated rotameter, which gave rise to a maximum superficial gas velocity of 0.75 m/s in the bed. In this regard, different superficial gas velocities were used for each system and temperature tested, covering both the fixed bed state and bubbling regime.

To investigate the effect of IPFs on the fluidization behavior, two systems were studied, uncoated sugar beads and coated sugar beads. Experiments of uncoated sugar beads were carried out at 20°C while the ones for coated sugar beads were conducted at different operating temperatures, 20°C, 30°C and 40°C. Hereafter, for simplicity, we name these systems with their different operating conditions in abbreviated form, SB, CSB20, CSB30 and CSB40, which stand for uncoated sugar beads at 20°C and coated sugar beads at 20°C, 30°C and 40°C, respectively. All experiments were performed at atmospheric pressure. It is worth noting that variations in the air density and viscosity in the 20°C to 40°C temperature range are 6% and 5%, respectively, which are fairly negligible compared to the amount of variation of cohesion, which rises from the new technique for the same temperature range. Moreover, the same amount of material was poured into the bed for both systems, which resulted in an initial bed height of approximately 20 cm at ambient conditions.

The pressure drop across the bed was measured using a differential pressure transducer. It was mounted flush with the wall of the bed through 15 micron inline filters and measuring ports. In addition, a local measurement technique was employed to investigate the influence of IPFs on the hydrodynamics of a gas-solid fluidized bed. To study the dynamic local flow structure a parallel optical fiber probe
was employed to measure the instantaneous local bed voidage. The probe was located 16 cm in height above the distributor and at the bed center for all the experiments. This axial position for the probe ensured it was far away from the turbulent effects of the distributor. For every temperature tested, the optical fiber probe was calibrated according to the linear interpolation between the read voltage of the air alone ($\varepsilon = 1$) and the read voltage of the fixed bed.

**Analysis Methods**

The ratio between the measured and calculated pressure drop across the bed, $\Delta P_m/\Delta P_c$, was used to highlight changes in the fluidization behavior of the coated sugar beads with increasing temperature.

Analysis of local bed voidage can effectively provide much information about the dynamics of the fluidized beds, which results in a better understanding about the flow behavior in the fluidized bed (8). In this regard, the dynamic local two-phase flow structure was analyzed for beds of different amounts of IPFs at each superficial gas velocity. Instantaneous local bed voidage was at first scrutinized to have a closer picture on gas and solids interaction in the bed for different operating conditions. The time-averaged local bed voidage at the same measurement position was also used to indicate the influence of IPFs on the local flow structure. The probability density function of the local voidage from $\varepsilon_{mf}$ to 1 was analyzed to quantitatively explain the gas-solid distribution of emulsion and dilute phases and its dependence on the IPFs. According to Cui et al. (9), this is allowed for investigating the dynamic behavior of the dilute and dense phases.

**RESULTS AND DISCUSSION**

First and foremost, it was necessary to confirm that the new methodology for the increment of cohesive IPFs can work properly after a slight increase in the system temperature. It was verified in the spheronizer where coated particles were prepared. To execute it, a required amount of inert particles with pharmaceutical excipients was first produced and, subsequently, coated with a 30$\mu m$ layer of the copolymer. Figure 1 shows the modification of the particulate system with temperature. It can be easily demonstrated that by progressively increasing the temperature, the surface of the particulate media, which was characterized by a smooth profile with no observable deviations or clusters relative to the mean position of the interface at 27°C, was gradually changed. It started to create apparent clusters at 36°C, which became more obvious at 39°C, followed by the appearance of significant structure modification at 40°C, the presence of a secondary particle flow structure at the top of the main particle bed at 41°C and, finally, the moving of the particle bed in mass at 43°C.

The most common approach for the overall identification of the fluidization status at different velocities is studying the whole bed pressure drop as a function of superficial gas velocity. The $\Delta P_m/\Delta P_c$ ratio can be used as a first indication of the effect of IPFs on fluidization behaviour (10). Figure 2 shows that by increasing IPFs, the degree of overshooting in the “fluidization” curve increases, which is accompanied by the increment of minimum fluidization velocity ($U_{mf}$). For the case of CSB40, IPFs considerably influenced the bed’s behavior and caused the presence of a mass of particles, which lifted as a plug rather than fluidizing. This effect is shown in Figure 2.
Furthermore, a decrement of the measured/theoretical pressure drop ratio from unity when the bed is fluidized shows the growth of the degree of cohesiveness of the bed (1, 10). As can be found in Figure 2, for CSB20 the $\Delta P_m / \Delta P_c$ ratio is similar to SB, while a decrement in the ratio is fairly observable for CSB30. For CSB40 fluidization was not attained due to paramount effect of cohesive IPFs. These results show that enhancement of IPFs can cause the behavior of the bed to change from Group B to Group A and even Group C powders.

Formisani et al. (11) noted that voidage of the loosely settled bed of particles of Groups A and B increases with temperature. They attributed these findings to the increment of IPFs with temperature. Table 3 reports the variation of fixed bed height with IPFs in this study. It reveals that the fixed bed height and, correspondingly, the fixed bed voidage increase with IPFs. This result affirms Formisani et al. (11) and indicates that a bed with higher IPFs can hold more gas in the fixed state.

The variations of local bed voidage with time reflect the interaction between gas and solids phases, which can influence the mass and heat transfer rates in the fluidized beds and, consequently, can affect the overall reaction rate in fluidized reactors (12). Figures 3 – 6 show the evolution of instantaneous local bed voidage with superficial gas velocity and IPFs for SB and CSB40. As can be found in these figures the portion of the dilute phase increases with increasing gas velocity for both cases. Therefore, the fluidizing air prefers to pass through the bed as a bubble phase. However, it seems that IPFs have the opposite influence on local bed behavior. It can be seen from Figures 3 – 6 that by increasing IPFs, the optical probe spends
more time in the emulsion phase rather than the dilute phase, which means that the portion of the emulsion phase increases by the enhancement of IPFs. As a result, the tendency of gas to pass the bed in the emulsion phase increases. It is worth noting that the CSB40 has a higher saturated emulsion value ($\varepsilon_{mf}$) compared to SB, which is a subsequent consequence of holding more gas in the fixed bed state. Moreover, a bed of higher IPFs has a dilute phase with a maximum voidage value lower than unity, while in the case of SB there are some temporal signals of bed voidage equal to unity, which are indications of pure bubbles. This can be referred to the presence of more solid in the dilute phase for the particulate bed of enhanced cohesive IPFs.

![Figure 3. Signal of instantaneous local bed voidage for SB (Ug = 0.41 m/s).](image3.jpg)

![Figure 4. Signal of instantaneous local bed voidage for CSB40 (Ug = 0.41 m/s).](image4.jpg)

![Figure 5. Signal of instantaneous local bed voidage for SB (Ug = 0.74 m/s).](image5.jpg)

![Figure 6. Signal of instantaneous local bed voidage for CSB40 (Ug = 0.74 m/s).](image6.jpg)

The time-averaged local bed voidage was further investigated to give valuable information about the effect of IPFs on the hydrodynamics of a gas-solid fluidized bed. As it is illustrated in Figure 7 the time-averaged local bed voidage increases with superficial gas velocity for particulate beds with different amounts of IPFs. Moreover, for each superficial gas velocity tested in the bubbling regime the time average local bed voidage decreases with increasing IPFs. This implies that the two-phase flow structure at constant superficial gas velocity varies with a greater extent for beds with higher IPFs.

Analysis of the probability density function of dynamic local voidage demonstrates that for each superficial gas velocity tested, the probability of the dense phase and dilute phase increases and decreases, respectively by increasing IPFs (Figures 8-10). These findings indicate that the probability of the flow structure with higher voidage is reduced and the whole flow structure becomes less dilute when IPFs are enhanced. In addition, it can be found that at a given fluidizing gas throughput, with
enhancing IPFs, more gas enters into the emulsion phase and dilutes it rather than passing into the bubble phase and increasing its fraction. Therefore, it can be inferred that the behavior of the particulate bed shifts from Group B into Group A by increasing IPFs in the bed.

Overall, the experimental results of this work are in good agreement with those for Yates and Newton (13) and Rowes et al. (14), who found that increasing the fines (< 45 μm) content results in an increase in interstitial gas flow. In the range of IPFs studied in this work, which defluidization phenomenon was not encountered within, the presence of more gas in the fixed bed state and emulsion phase is an indication of the improvement of gas/solid contact in the bed of Group B powders.

**CONCLUSION**

This work introduces a new and different approach to study the effect of IPFs on the fluidization behavior of the gas-solid fluidized bed. It was shown that by controlling the temperature of the particulate system of inert particles coated with a layer of a PMMA/PEA copolymer, the degree of cohesiveness of the bed can be easily changed in a selective manner. Interesting aspects of this approach for increment of IPFs are working at low bed temperatures, controlling the level of cohesiveness by merely controlling the bed temperature and facilitating the investigation of the influence of IPFs on fluidization characteristics from low to high superficial gas velocities.

Experimental results obtained by local and global measurement techniques show that increment of IPFs can significantly affect the dynamic local flow structure and overall behavior of the gas-solid fluidized bed. Dictating higher amounts of IPFs into
the bed causes higher fixed bed voidage and minimum fluidization velocity. Similarly, there is a significant influence of IPFs on the bed behavior in the bubbling fluidization regime. The results obtained in the range of IPFs studied in this work imply that at a given throughput of the fluidizing gas, the relative tendency of gas flowing in the emulsion phase increases with IPFs, which has a positive effect on enhancing chemical conversion in the case of the active catalyst. According to experimental results, it can be concluded that by increasing IPFs the fluidization behavior of the particulate bed can shift from Group B to Group A or even Group C powders.

NOTATION

<table>
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<th>Variable</th>
<th>Description</th>
<th>Symbol</th>
<th>Unit</th>
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</thead>
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<td>$A$</td>
<td>bed cross-section area (m$^2$)</td>
<td>$A$</td>
<td>m$^2$</td>
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<tr>
<td>$d_p$</td>
<td>mean particle diameter ($\mu$m)</td>
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<td>$\mu$m</td>
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<tr>
<td>$M$</td>
<td>bed weight (kg)</td>
<td>$M$</td>
<td>kg</td>
</tr>
<tr>
<td>$U_{mf}$</td>
<td>minimum fluidization velocity (m/s)</td>
<td>$U_{mf}$</td>
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<td>$U_g$</td>
<td>superficial gas velocity (m/s)</td>
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<tr>
<td>$\Delta P_m$</td>
<td>measured pressure drop (N/m$^2$)</td>
<td>$\Delta P_m$</td>
<td>N/m$^2$</td>
</tr>
<tr>
<td>$\rho_p$</td>
<td>particle density (kg/m$^3$)</td>
<td>$\rho_p$</td>
<td>kg/m$^3$</td>
</tr>
</tbody>
</table>

Greek Letters

- $\varepsilon$: local voidage (-)
- $\varepsilon_{mf}$: minimum fluidization voidage (-)

REFERENCES