SEGREGATION IN DENSE GAS_FLUIDISED BEDS:
VALIDATION OF A MULTI-FLUID CONTINUUM MODEL WITH
NON-INTRUSIVE DIGITAL IMAGE ANALYSIS MEASUREMENTS

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ABSTRACT

A non-intrusive digital image analyses technique is applied to study size driven segregation of a binary mixture of coloured glass beads in a bubbling gas-fluidised bed. Segregation rates and patterns obtained from experiments are compared to numerical simulations performed with a two-dimensional multi-fluid Eulerian model, that uses closure laws according to the kinetic theory of granular flow. It is demonstrated that prediction of segregation is a rather severe test case for fundamental hydrodynamic models, since bubble dynamics and momentum transfer between particles of different classes have to be modelled correctly. At all gas velocities segregation rates predicted by the multi-fluid model were much higher than those observed in experiments. At gas velocities higher than the minimum fluidisation velocity of the largest particles the model still predicts segregation, when it does not occur in experiments. It is concluded that the predicted intensity of bubbling is too low, since energy dissipation by particle-particle interaction is still underestimated in the applied kinetic theory closure model.

INTRODUCTION

In many industrial dense gas-fluidised bed processes, e.g. gas-phase polymerisation and fluidised bed granulation, mixtures of particles with different physical properties are encountered. When particles differ in size and/or density, segregation may occur. Segregation is most marked at low gas velocities when there is appreciable particle density difference. Even a strongly segregating system, however, can be fairly well mixed if the gas velocity is increased sufficiently although it can be difficult to remove the last traces of segregation.

Much has been reported on particle mixing and segregation in dense gas-fluidised beds (1-3). Bubbles have been indicated as the vehicle for both mixing and segregation. Rising bubbles carry a mixture of particles to the top of the bed and disturb the packing state of the bed so that segregation of larger or heavier particles may occur. Up to now most authors only report steady-state axial segregation profiles and hardly any information can be found on the spatial distribution and rate of segregation (4,5). Combined measurements of segregation and bubble dynamics performed with particles with well defined properties (size, shape, density and collision properties) in a bed with a well defined geometry and gas inflow on short time scales that can currently be handled by fundamental hydrodynamic models (CFD-models) are not yet available. However, for thorough validation these models this kind of information is required and therefore a digital image analyses technique has been developed to study segregation of mixtures of coloured particles in transparent pseudo two-dimensional gas-fluidised beds. A brief outline of this technique and some experimental results will be presented in this paper.

Accurate prediction of segregation is required to improve the design, operation and scale-up of gas-fluidised bed processes. In the last decade considerable progress has been made in the area of fundamental hydrodynamic modelling of gas-fluidised suspensions and fluidised beds.
can nowadays be modelled with discrete particle Lagrangian and multi-fluid Eulerian models. Hoomans et al. (6) demonstrated the ability of discrete particle models to predict segregation in dense gas-fluidised beds. They observed a strong influence of the particle-particle collision parameters on bubble dynamics and consequent segregation rates. Mathiessen et al (7) successfully modelled axial segregation patterns obtained with LDA and PDA techniques in a laboratory scale riser, using a multi-fluid model. In riser flows kinetic momentum transfer forms the dominant momentum transfer mechanism. In this work we will apply a multi-fluid model to model segregation in a dense gas-fluidised bed, where particle-particle interactions dominate momentum transfer.

DIGITAL IMAGE ANALYSIS

Agarwal et al. (5) demonstrated that non-invasive digital image analyses techniques can be applied to study bubble dynamics, mixing and segregation in pseudo two-dimensional bubbling gas-fluidised beds. In this work experiments with binary mixtures of coloured glass beads were performed in a pseudo two-dimensional bubbling fluidised bed. The experiments were recorded at a rate of 25 frames/second by a 3-CCD colour camera and stored on a harddisk by a framegrabber. A digital image analysis technique was developed to correlate the measured light intensities to bubble patterns and particle volume fractions.

Experimental setup

The experiments were carried out in a 15 cm wide, 70 cm high, 1.5 cm deep pseudo two-dimensional gas-fluidised bed constructed of glass. Visual observation of the lowest 1.3 cm of the bed close to the gas distributor is obstructed by a flange, which is applied to mount the bed to the distributor. A continuous, high intensity, uniform illumination of the bed is obtained with six 500 Watt halogen lamps which are mounted next to the bed. Light emitted by these lamps is projected on the bed by two diffuse reflectors. A blue background that gives good contrast with the colours of the particles is placed behind the bed to improve bubble detection.

To obtain a severe test case for fundamental hydrodynamic models a binary mixture of coloured glass beads that differed less than a factor 2 in size was taken. Initially the bed was filled up to 15 cm with a 50/50 volume percent mixture of glass beads. A well-mixed initial condition was obtained by fluidising the particles at a velocity somewhat higher than the minimum fluidisation velocity of the largest particles for several minutes, where after the gas-inflow was suddenly shut down. The particle properties of the spherical glass beads that were used are summarised in table I. Minimum fluidisation velocities were determined experimentally from pressure drop measurements over the bed and particle collision parameters were obtained from detailed impact measurements performed by the Impact Research Group of the Open University at Milton Keynes.

<table>
<thead>
<tr>
<th>Table I: Properties of glass beads</th>
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</thead>
<tbody>
<tr>
<td>small particles</td>
</tr>
<tr>
<td>large particles</td>
</tr>
<tr>
<td>colour: yellow</td>
</tr>
<tr>
<td>diameter, d</td>
</tr>
<tr>
<td>density, ρ</td>
</tr>
<tr>
<td>minimum fluidisation velocity, u_{mf}</td>
</tr>
<tr>
<td>coefficient of normal restitution, e</td>
</tr>
<tr>
<td>coefficient of friction, μ</td>
</tr>
<tr>
<td>coefficient of tangential restitution, β₀</td>
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**Analysis procedure**

The recorded images consist of 576x720 picture elements (pixels) that contain information about the local red, green and blue light intensity levels. The blue background emits mainly blue light and the red particles emit mainly red light. The yellow particles emit both green and red light and the absolute red intensity emitted by the yellow particles is even higher than that emitted by the red ones. The bed is placed in the middle of the picture and covers about 129 x 531 pixels, so the pixels are slightly smaller than the particles.

In the first step of the analysis bubbles are detected at pixel level. The high blue intensity level of the background is used to detect large bubbles that span the whole depth of the pseudo two-dimensional bed. Small bubbles that can be observed as shaded areas are removed based on their relatively low red intensity level, which is lower than the red intensity level emitted by the red as well as the yellow particles.

In the second step of the analysis the composition of the particle mixture outside the bubbles is determined. To obtain interrogation areas that contain a sufficient amount of particles to perform concentration measurements, colour intensity information of pixels that are not assigned to bubbles is combined on a 1 cm x 1 cm mesh. The ratio of the red and the green light intensity is then used to determine the mixture composition in each element of the mesh (cell). Therefor a calibration curve has been recorded with 11 mixtures of known composition in a well-mixed packed bed. Using this calibration curve the particle concentrations could be measured everywhere within the fluidised bed with an accuracy of approximately 10%. An example of the analysis of a single image is given in figure 1.

**Experimental results**

The segregation pattern and rate of the binary mixture of glass beads was studied at three different gas velocities, 1.10 m/s, 1.25 m/s and 1.40 m/s. At 1.10 m/s, which is in between the minimum fluidisation velocity of both components, rapid segregation was observed. The segregation patterns given in figure 2 show that within 15 seconds the whole top layer of the bed is occupied by small particles. Within the first minute the top layer of small particles grows significantly, while two heaps of (defluidised) large particles form near the bottom. The segregation rate has been quantified with the digital image analysis technique. Therefor the...

![Figure 1: Results of digital image analysis of a single image taken from an experiment at 1.10 m/s at t = 15.36 s. High grey intensities indicate high phase volume fractions.](image)
Figure 2: Snapshots of a segregation experiment at 1.10 m/s.

Figure 3: Average particle heights obtained from digital image analysis of a segregation experiment at 1.10 m/s.

Figure 4: Average particle heights obtained from digital image analysis of a segregation experiment at 1.25 m/s.

Figure 5: Snapshot of a segregation experiment at 1.25 m/s at t = 60.00 s.
average height of both particle phases in the bed was calculated from
\[
\langle h_k \rangle = \frac{\sum_{\text{Cells}} \varepsilon_{k,\text{cell}} h_{\text{cell}}}{\# \text{Cells}},
\]
whereby \( \varepsilon_{k,\text{cell}} \) is the volume fraction of particle phase \( k \) in the cell and \( h_{\text{cell}} \) the height of the cell. The results of the analysis, given in figure 3, show that the segregation rate is high during the first 40 seconds, where after segregation is nearly complete and the dynamic equilibrium with mixing is slowly approached. Further it can be seen from the enlargement of the first 10 seconds of the experiment that the bed expansion frequency (= large bubble frequency) is about 1.8 Hz and the amplitude of the average bed expansion is about 0.9 cm.

At 1.25 m/s, the minimum fluidisation velocity of the largest particles, some segregation takes place near the top and the bottom of the bed, but these segregated regions get quickly mixed up with the well-mixed bed content by bubbling of the bed. Figure 4 shows that segregation is hardly traceable during the first 60 seconds. The bed expansion frequency is about 2.0 Hz and the amplitude of the average bed expansion is about 1.2 cm. A snapshot of the bed taken after one minute of fluidisation is shown in figure 5.

At 1.40 m/s bubbling is even stronger and segregation is hardly traceable. The bed expansion frequency is about 1.9 Hz and the amplitude of the average bed expansion is about 2.4 cm.

**MULTI-FLUID MODELLING**

The segregation process has been modelled with a multi-fluid Eulerian model. In this type of model a particle mixture is divided into a discrete number of classes, whereby different physical properties can be specified for each class. The model uses closure equations derived from the kinetic theory of granular flow to describe particle-particle interactions. Several authors have worked on the derivation of these constitutive equations for binary and multi-component mixtures (8-11). The multi-fluid model and the kinetic theory closure equations applied in this work can be found in (12).

**Frictional viscosity**

The kinetic theory only accounts for kinetic and collisional contributions to the viscosity of the particulate phases. In the quasi-static regime close to or below minimum fluidisation, however, the dominant stress generation mechanism is due to long-term particle-particle contacts. Laux (13) tested several frictional viscosity models from the field of soil mechanics and demonstrated that it is worthwhile to include such a frictional stress model into Eulerian continuum models to improve the description of phenomena such as heap formation upon defluidisation. The frictional viscosity model that gave the best results is given by
\[
\mu_{k,\text{fric}} = \frac{6 \sin \phi_f}{9 - \sin^2 \phi_f} \left| \frac{I_{II}}{2 \sqrt{3} I_{II_D}} \right|
\]
where \( \phi_f \) is the internal angle of friction (45° in this work), \( I_{II} \) is the first invariant of the particle pressure stress tensor and \( I_{II_D} \) is the second invariant of the deviator of the rate-of-strain tensor. It is further assumed that long-term particle contacts only contribute to the stress tensor above a certain particle volume fraction \( \varepsilon_{\text{im}} \) (0.5 in this work). The effective hydrodynamic viscosity should then be given by the maximum of the viscosity obtained from the kinetic theory model and the frictional viscosity model.
\[
\mu_{k,\text{eff}} = \begin{cases} 
\max \left\{ \mu_{k,\text{TGF}}, \mu_{k,\text{fric}} \right\} & \varepsilon_k \geq \varepsilon_{\text{lim}} \\
\mu_{k,\text{TGF}} & \varepsilon_k \ < \varepsilon_{\text{lim}}.
\end{cases}
\]

Finally, to guarantee numerical stability and reasonable convergence rates the viscosity is limited to \( \mu_{\text{lim}} (1 \times 10^3 \text{ Pa}\cdot\text{s} \) in this work)
\[
\mu_k = \min \left( \mu_{k,\text{eff}}, \mu_{\text{lim}} \right).
\]

Packing density of mixtures

Blending of particles of several sizes will lead to an increase in the maximum packing density of the bed. Fedors and Landel (14) found that a good approximation of experimental data for the maximum random packing density of a binary mixture of two components \( k \) and \( l \) of different size \( d_k > d_l \) as a function of the mixture composition \( X_k \) \((= \varepsilon_k / (\varepsilon_k + \varepsilon_l))\) is given by:
\[
\varepsilon_{kl}^\text{max} = \left( \varepsilon_k^\text{max} - \varepsilon_l^\text{max} + \left(1 - \frac{d_l}{d_k}\right) \left[1 - \varepsilon_k^\text{max} \varepsilon_l^\text{max} \left(1 - X_k\right) + \varepsilon_k^\text{max} \right] \right) X_k + \varepsilon_l^\text{max} \quad X_k \leq \frac{\varepsilon_k^\text{max} + \left(1 - \varepsilon_k^\text{max} \varepsilon_l^\text{max} \right)}{\varepsilon_k^\text{max} + \left(1 - \varepsilon_k^\text{max} \varepsilon_l^\text{max} \right)}
\]
\[
\varepsilon_{kl}^\text{max} = \left(1 - \frac{d_l}{d_k}\right) \varepsilon_k^\text{max} + \left(1 - \varepsilon_k^\text{max} \varepsilon_l^\text{max} \right) \left(1 - X_k\right) + \varepsilon_l^\text{max} \quad X_k \geq \frac{\varepsilon_k^\text{max} + \left(1 - \varepsilon_k^\text{max} \varepsilon_l^\text{max} \right)}{\varepsilon_k^\text{max} + \left(1 - \varepsilon_k^\text{max} \varepsilon_l^\text{max} \right)}
\]

whereby \( \varepsilon_k^\text{max} \) and \( \varepsilon_l^\text{max} \) are the maximum particle packing fractions of the single components of the mixture \((0.64356 \text{ in this work})\).

In the kinetic theory of granular flow the radial distribution function has to account for the dependence of the maximum particle packing density on the local mixture composition. Jenkins and Mancini (8) applied the radial distribution of Mansoori et al. (15), since it was in best agreement with numerical simulations of a mixture of hard spheres. For binary mixtures it can be written as:
\[
g_0(\varepsilon_k, \varepsilon_l, d_k, d_l) = \frac{1}{1 - \varepsilon_k - \varepsilon_l + 3 \frac{d_l \varepsilon_k + d_k \varepsilon_l}{(d_k + d_l)(1 - \varepsilon_k - \varepsilon_l)^2} + 2 \left( \frac{d_l \varepsilon_k + d_k \varepsilon_l}{(d_k + d_l)^2 (1 - \varepsilon_k - \varepsilon_l)^3} \right)^2}.
\]

However, this radial distribution function does not approach infinity near the maximum packing density of the mixture and application of it to dense gas-fluidised bed simulations will lead to prediction of too high particle packing densities. Therefore the maximum packing fraction that appears in the radial distribution function which is generally applied in dense gas-fluidised bed simulations was presumed to be a function of the mixture composition, leading to
\[
g_0(\varepsilon_k, \varepsilon_l, d_k, d_l) = \left(1 - \left( \frac{\varepsilon_k + \varepsilon_l}{\varepsilon_{\text{max}}^{\text{kl}}} \right)^{1/3} \right)^{-1},
\]

whereby the maximum packing fraction is taken from the equations by Fedors and Landel (14).

Modelling results

All simulations were carried out on a Cartesian grid with 30 x 80 elements of 0.5 x 0.5 cm. The second order Barton scheme (16) was applied to calculate the convective fluxes. Initially particles of both sizes were well mixed and at \( t = 0.0 \) the inflow velocity was stepwise set to the required fluidisation velocity. To obtain more realistic start-up behaviour, small random perturbations of at most 1% were applied to the initial volume fractions to remove symmetry.
The Ergun equation, which is applied to describe gas-particle drag in the dense fluidised bed, predicts somewhat higher values for the minimum fluidisation velocity of both components (small particles: \( U_{mf} = 0.92 \) m/s; large particles: \( U_{mf} = 1.28 \) m/s) compared to the experimental findings. Therefore the numerical simulations were performed at slightly higher velocities than the experiments, namely at 1.15 m/s, 1.30 m/s and 1.45 m/s.

The average particle heights predicted by the model are shown in figure 6. Comparison to figures 3 and 4 clearly shows that the model predicts too high segregation rates. Further strong segregation is predicted at and above the minimum fluidisation velocity of the largest particles, when segregation does not occur in experiments. Snapshots from the simulation at 1.30 m/s (given in figure 7) show that large particles are transported to the top of the bed in the wake of a bubble. But, as can be seen from figure 6, apart from the initial bed expansion caused by the start-up of the simulation, the amplitude of the bed expansion in the simulations is far smaller than in the experiments. So the model underestimates the bubble intensity and therefore mixing.

In the simulation at 1.15 m/s, below the minimum fluidisation velocity of the large particles, the formation of a stagnant zone with heaps of large particles near the bottom of the bed was not reproduced. This is probably due to a too low limiting value for the shear viscosity \( 10^{3} \) Pa·s.

![Figure 6](image.png)

**Figure 6:** Average particle heights obtained from multi-fluid continuum simulations

![Figure 7](image.png)

**Figure 7:** Snapshots of the segregation pattern obtained by a simulation at 1.30 m/s
DISCUSSION AND CONCLUSION

The sensitive dynamic equilibrium between segregation and mixing of a binary mixture of particles that differ less than a factor 2 in size was studied to obtain a severe test case for fundamental hydrodynamic models. Segregation rates and patterns obtained from non-intrusive digital image analysis measurements were compared to simulation results obtained with a multi-fluid continuum model. It is demonstrated that a multi-fluid model can be used to study segregation in dense gas-fluidised beds, though with the current 'state-of-the-art' kinetic theory closure equations, the predicted segregation rates are too high.

Since bubbles play a dominant role in both segregation and mixing, correct prediction of bubble behaviour is essential. It was observed that the bubble intensity in the simulations was much lower than in the experiments. In earlier work (12) we concluded that bubble dynamics in dense gas-fluidised beds strongly depend on the amount of energy dissipated by particle-particle collisions. In the current kinetic theory closure equations only energy losses due to restitution are taken into account, though for Geldart B and D-type particles a large amount of kinetic energy is transferred to rotation and dissipated by friction. Another shortcoming of the current kinetic theory of granular flow closure equations is that structure formation and collective motion that may occur close to the densest particle packing are not accounted for.

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REFERENCES