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Refereed Proceedings

2013

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Recommended Citation

Alberto Di Renzo, Rossella Girimonte, Vincenzino Vivacqua, and Francesco P. Di Maio, "Experimental Verification of the Particle Segregation Model Predictions for Fluidized Biomass/Inert Mixtures" in "The 14th International Conference on Fluidization – From Fundamentals to Products", J.A.M. Kuipers, Eindhoven University of Technology R.F. Mudde, Delft University of Technology J.R. van Ommen, Delft University of Technology N.G. Deen, Eindhoven University of Technology Eds, ECI Symposium Series, (2013). http://dc.engconfintl.org/fluidization_xiv/107

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EXPERIMENTAL VERIFICATION OF THE PARTICLE SEGREGATION MODEL PREDICTIONS FOR FLUIDIZED BIOMASS/INERT MIXTURES

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ABSTRACT

Segregation in bubbling fluidized binary mixtures is studied at low velocities by comparing experimental observations of the concentration profiles with predictions of the segregation direction obtained by the recently-proposed Particle Segregation Model (PSM, <u>11</u>) for biomass/inert systems. Biomass/glass beads and biomass/sand pairs with different inert sizes are investigated deriving, for each one, the component distribution along bed height at various volume fractions. Remarkably, segregation direction reversal with bed composition is obtained for all systems, as prescribed by the PSM, although quantitative discrepancies are observed between the actual and predicted equilibrium compositions.

INTRODUCTION

One of the advantages of the bubbling fluidized bed technology is the ability to provide good solids mixing in multicomponent systems, e.g. in reactor systems. However, the level of bed homogeneity may be insufficient, particularly at low velocities, where the bubble-induced agitation is not high enough to guarantee solids mixing: the solids tend to stratify and segregation issues are faced (<u>1</u>).

Regarding the tendency to segregation of binary fluidized beds, solids properties as well as the gas velocity are known to be the main variables affecting the process. In the literature, several works report attempts to characterize the behavior of mixtures. However, the problem of predicting the segregation direction and intensity as a function of the components' and bed properties remains largely unsolved, particularly with irregularly shaped solids like biomass particles. A significant number of experimental works have focused on the individual effect of size segregation and density segregation, or a combination of both, see e.g. (2-4). More recently, numerical simulations have supplemented these (5-7) to examine the process at a smaller scale.

The traditional nomenclature of *flotsam* and *jetsam*, introduced by Rowe et al. (8), will be adopted. With reference to the segregation behavior of fluidized binary mixtures, one of the earliest distinguishing criteria was proposed by Chiba et al. (9), based on their experimental observations. Recently, Rao et al. (10) produced a global, empirical classification based on a significant number of data from the literature and their own. From the theoretical point of view, a relatively simple framework for predicting the segregation direction of ideal mixtures has been developed by the authors (11), as summarized in Section "The Particle Segregation Model (PSM)". By segregation direction, the tendency to act as flotsam of one or the other of the two bed constituents is intended here. Despite good agreement was observed with published data (11), additional experimental validation of the PSM predictions in real applications is required. In particular,

one simple application of the PSM allows imposing segregation equilibrium at a given composition and deriving one of the four solid properties that define the size ratio and the density ratio. As a result, segregation direction reversal is predicted to occur for compositions higher and lower than that imposed for equilibrium. Therefore, in the present work, experiments involving a mixture of biomass particles and inert solids of specific sizes are carried out, aiming at verifying the conditions for the segregation direction reversal to occur.

THE PARTICLE SEGREGATION MODEL (PSM)

A short account of the particle segregation model (PSM) result, discussed in details in Ref. (<u>11</u>), is reported here. The PSM is based on an elementary concept: the mechanical equilibrium is applied to one particle in the binary mixture. Thus, a comparison is analytically derived between the hydrodynamic force N, pushing the particle upwards, and the gravitational force G, pulling the particle downwards. Expressed as the ratio of the two terms, equilibrium is obtained when

$$\frac{N}{G} = 1 \tag{1}$$

The most complex task is the correct formulation of the hydrodynamic force N exerted on a particle immersed in a binary fluidized bed. A general and convenient expression of the inter-phase momentum transfer is through the subdivision of the total force in a generalized buoyancy B, dependent on the pressure gradient, and a pure drag force F, dependent on the gas-particle relative velocity. In a fluidized bed, the former can generally be expressed in terms of the bulk density of the suspension, as:

$$B = -\nabla P \cdot V = \rho (1 - \varepsilon) g V \tag{2}$$

For the drag force F, various expressions have been proposed for a particle immersed in a polydisperse suspension. Among these, models formulated on the basis of Lattice-Boltzmann simulation results (<u>12</u>) and their extensions (<u>13-14</u>) are most appropriate. The point of departure of such derivation is the expression of the force acting on one particle species in terms of a coefficient multiplying an appropriate average force, i.e.:

$$F_i = y_i^2 \overline{F} \tag{3}$$

The average force is to be intended as the drag force acting on a particle of average diameter \overline{D} , while the dimensionless coefficient is the square of the polydispersion index y_i , defined as:

$$y_i = \frac{D_i}{\overline{D}} \tag{4}$$

The average diameter follows the well-accepted definition of Sauter's mean diameter:

$$\overline{D} = \left(\frac{x_1}{D_1} + \frac{x_2}{D_2}\right)^{-1}$$
(5)

The subsequent step is the calculation of the fluidization velocity, necessary to evaluate the drag force. The model is based on the assumption that bubbles are not dominating the fluid dynamics, so that the dense phase superficial velocity can be assumed equal to the minimum fluidization velocity of the suspension, and this is applied throughout the bed. Moreover, the bed is assumed fully mixed, in a way that the overall system composition equals the nominal fraction initially charged in the bed. The minimum fluidization velocity of such a bed is the value necessary to suspend a homogeneous bed of average density

$$\overline{\rho} = x_1 \rho_1 + (1 - x_1) \rho_2 \tag{6}$$

and size equal to the average diameter \overline{D} . Without loss of generality, we will assume species 1 to be the smaller component. Let us consider a particle of species 2 subjected to the action of the fluid flowing at the average minimum fluidization velocity \overline{u}_{mf} . Under the hypothesis of low-Re flows, it can be shown (see Ref. (<u>11</u>)) that application of Eq. (3) leads to the following expression:

$$F_2\Big|_{\overline{u}_{mf}} = \frac{1}{6}\pi g \varepsilon \overline{\rho} \overline{D} D_2^2 \tag{7}$$

Using Eqs (2) and (7), the equilibrium condition prescribed by Eq. (1) becomes:

$$V_2 \overline{\rho} (1 - \varepsilon)g + \frac{1}{6} \pi g \varepsilon \overline{\rho} \overline{D} D_2^2 = V_2 \rho_2 g$$
(8)

By introducing the particle property ratios:

$$s = \frac{\rho_2}{\rho_1} \tag{9}$$

$$d = \frac{D_1}{D_2} \tag{10}$$

and expressing the average density and diameter in terms of the single species properties, some manipulations allow formulating Eq. (8) as:

$$s = \frac{x_1}{\frac{1}{1 - \varepsilon + \varepsilon \cdot \left(\frac{x_1}{d} + 1 - x_1\right)^{-1}} - (1 - x_1)}$$
(11)

Typical biomass/inert mixtures are complex because bigger particles (biomass) are very often also lighter than inert solids (e.g. sand). In this regard, note that considering D_1 as smaller than D_2 , corresponding to ρ_1 larger than ρ_2 , leads to both property ratios, as defined in Eq. (9) and (10), that range from 0 to 1.



Figure 1. Segregation equilibrium lines (Eq. (11)) at the different compositions and segregation direction sketched with smaller-denser (dark gray) and larger-less dense (light gray) species. Voidage equals 0.4.

Combinations of properties satisfying Eq. (11) correspond to mixtures at mechanical equilibrium, i.e. systems that initially loaded in a fully mixed state would remain in that state. Derivations involving the inequality allow, without adjustable parameters, evaluating the segregation direction, i.e. showing that the smaller species will act as flotsam/jetsam for s values larger/smaller than the value prescribed by Eq. (11) (Fig. 1).

The assumption of minimum fluidization conditions limits the applicability of the PSM to low-velocity fluidized systems. On the other hand, PSM results prove useful in providing the inherent segregation tendency of mixed beds. It is the case to recall that bubbling beds at higher velocities also exhibit the minimum fluidization conditions (voidage and velocity) in the emulsion phase. Therefore, the segregation tendency prescribed by the PSM, at least as one of the driving forces, should apply also to commercial fully fluidized beds. Stronger limits do apply, instead, on the predictability of the final degree of mixing/segregation, which presently remains outside the scope of the PSM (11).

MATERIALS AND EXPERIMENTAL PROCEDURE

All experiments are carried out in a 10-cm diameter (9.3-cm ID) Perspex cylindrical fluidization column, equipped with a 4-mm-thick plastic porous distributor. Air is the fluidizing medium, whose flow from a compressor is regulated by a set of rotameters. Fluidization tests involving binary beds are all performed starting from an initially mixed bed. Bed heights are such that a height to diameter of 1.7 is obtained. Measurements of the concentration profiles in the column are obtained after "freezing" the bed (i.e. shutting the gas flow suddenly); then, horizontal layers of particles, about 1 cm thick, are withdrawn from the top of the column, separated by sieving and weighed.

The mixtures investigated involve different compositions of biomass particles and various inert particles. Biomass is available in form of dried, crushed olive pits (OP), while glass ballotini and sand are used as inert to represent typical industrial charges. The original biomass stock was highly polydisperse and had a relatively large size. Sieving in the range 1.4–2 mm was necessary to bring the sample within workable diameters. Owing to the uncertainties related to the irregular shape, a hydrodynamic determination of its average size through pressure drop measurements was performed, in combination with a digital image analysis for shape characterization. More details can be found in another paper

to appear in the conference proceedings (<u>15</u>). Inert particles (glass ballotini, GB, and two sizes of sand granules, SG) were available in different cuts, and their final sizes were selected in order for the resulting mixture to fall within the shaded area of Fig. 1, as described in the next subsection. Fig. 2 shows a photograph of the biomass and glass ballotini. The properties of the examined materials, including the experimentally determined u_{mfr} are listed in Table 1 and the resulting mixtures in Table 2.

Table 1	Properties	of the solids.
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Solid	Identifier	ho (kg/m ³)	<i>D</i> (mm)	u_{mf} (cm/s)
Olive pits (biomass)	OP1540	1380	1.54	66.1
Glass ballotini	GB250	2480	0.25	6.6
Sand granules	SG340	2590	0.34	22.8
	SG160	2590	0.16	5.6

Table 2	. Mixtures	investigated.
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	V		
Mixtures (inert-biomass)	$d = D_1 / D_2$	$s = \rho_2 / \rho_1$	equilibrium x_1 (Eq.11)
GB250-OP1540	0.16	0.56	0.51
SG340-OP1540	0.22	0.53	0.29
SG160-OP1540	0.10	0.53	0.57



Figure 2. Photograph of biomass particles (left) and glass ballotini (right).

A segregation reversal experiment

The PSM main result, represented by Eq (11), is the capability to predict the segregation direction based on the size ratio, density ratio and bed composition (at a given voidage value). Alternatively, one can decide to achieve a given segregation direction and derive one of the properties leading to the prescribed condition (i.e. bed composition, one density or one size). In particular, by selecting solids pairs whose size and density ratios fall within the shaded area of Fig. 1, the model predicts that for low compositions one species will act as flotsam, while for higher compositions the other component will be the flotsam. In other words, a change of segregation direction (reversal) is predicted to occur with bed composition. The occurrence of this peculiar phenomenon is used as test with specific experiments against the theoretical predictions of the PSM. All the mixtures reported in Table 2 fulfill the prescribed condition, with segregation equilibrium at compositions (i.e. the predicted point of reversal, assuming $\varepsilon = 0.4$) also reported in Table 2. Note that the mixtures exhibit a change of voidage with composition that has typically very significant implications, e.g. on u_{mf} . However, as discussed in (11), the effects on the PSM predictions are rather limited and, considering the degree of uncertainty in other properties (the equivalent diameters above all), computations are retained at the reference voidage of 0.4.

RESULTS AND DISCUSSION

The main aim of the present analysis is the verification of the peculiar behavior of the segregation direction reversal. This task is carried out in both qualitative and quantitative terms, i.e. the occurrence of the phenomenon, which – it is to remark – has never been reported and investigated in the literature, is checked first; then the corresponding bed composition is compared with the PSM prediction.

Since the particle segregation model applies under the hypothesis of gas velocities just above the minimum fluidization condition, concentration profiles were obtained at velocities laying halfway between the initial and final fluidization velocities (<u>15</u>), at each composition. Each test, repeated twice and in a few cases three times for the biomass-sand mixtures, required waiting several minutes to ensure steady-state, as visually observable.

Measurements of the volumetric fraction profiles of the inert components along bed height are reported in Figs. 3-5. In all plots, the actual profiles are reported together with the nominal compositions. The distribution of the concentrations with bed height allows the segregation direction to be qualitatively assessed and the reversal to be identified quantitatively. It is observed that a segregation direction inversion is indeed found for all the three systems under scrutiny. For the GB250-OP1540 mixture, at nominal concentrations up to 0.3 there is a clear prevalence of the glass ballotini in the upper portion of the bed (Fig. 3). On the contrary, for glass ballotini volume fractions of 0.5 or higher, it is the biomass that prevails in the upper layers of the fluidized bed, acting as flotsam. For a volumetric fraction of 0.4, substantially mixed layers are found throughout the bed. The mixture composed by biomass particles and the bigger cut of sand (SG340-OP1540) exhibits a segregation reversal as well. In this case, for volume fractions up to 0.6 the inert particles appear as flotsam, while starting from 0.7 the biomass tends to float to the surface, with the profiles showing a quite peculiar "belly" corresponding to about half the bed height. Note that repeated experiments have confirmed such behavior. In any case, for both GB250-OP1540 and SG340-OP1540 mixtures the extent of segregation in the bed was relatively modest. The use of finer sand granules as inert, bringing the bigger-tosmaller size ratio up to about ten (i.e. with solid-solid percolation), leads to a stronger tendency towards segregation. For the SG160-OP1540 mixture, the fine sand tends to stratify to the top of the bed right from the lowest compositions investigated, creating a nearly pure layer at volume fractions between 0.4 and 0.6. At higher values, the upper layer shows a slight decrease of the fraction of fines, and the upper portion of the profile shows an inverted profile, with a decreasing fraction of sand particles with bed height. This result is more clearly observable at $x_1 = 0.8$. Although it is less than a clear manifestation of a direction reversal, it is quite evident that a change of segregation pattern occurs at the higher volume fractions with respect to the lower values also for this system. The analysis is complicated by the fact that bed fluidization occurs starting from the bottom of the bed instead of from the top. As soon as the bed is suspended, the upper part of the bed is lifted by the fluidizing lowermost section of the bed.

Considering the number and significance of the approximations in the derivation, the very simple mathematical form of the particle segregation model (and the lack of fitting parameters), it is remarkable that the predictions of segregation reversal, at least for the systems investigated, are quite confirmed. In quantitative terms, for the biomass coupled to glass ballotini a relatively good agreement is found between the predicted equilibrium ($x_1 = 0.51$) and the observed value ($x_1 \approx 0.4$) (Fig. 3). On the other hand, with the bigger cut of sand particles segregation

equilibrium occurs for the inert-rich mixtures rather than the predicted biomassrich systems (observed equilibrium $x_1 \approx 0.65$ vs. predicted equilibrium $x_1 = 0.29$) (Fig. 4). With finer sand, a shift of the equilibrium towards higher volume fractions is observed, as predicted by the PSM, but with higher values of the composition (Fig. 5).



Figure 3. Glass ballotini concentration profiles along bed height at different overall volume fractions for the GB250-OP1540 mixture.



Figure 4. Sand granules concentration profiles along bed height at different overall volume fractions for the SG340-OP1540 mixture.



Figure 5. Sand granules concentration profiles along bed height at different overall volume fractions for the SG160-OP1540 mixture.

CONCLUSIONS

The problem of characterizing the segregation tendency of binary mixtures of practical interest was studied both theoretically and experimentally. The recently proposed particle segregation model (PSM), based on the concept of force balance on a particle immersed in a binary bed at minimum fluidization, was shortly summarized. Experiments were carried out on mixtures of biomass combined to three inert solids (glass ballotini e two sand cuts). Based on the model predictions all mixtures should exhibit a segregation direction reversal with bed composition. Measurements of the composition profiles along bed height confirmed the presence of a segregation reversal with bed composition. Although the capabilities to predict the composition for segregation reversal quantitatively can be judged only satisfactory, the fact that a simple, first-principle and parameter-free model is able to capture the essence of a quite peculiar phenomenon for gas-fluidized beds represents a significant advancement of the theoretical tools available in the field and, possibly, it represents a good point of departure for more sophisticated and more accurate models.

NOTATION

- *B* generalized buoyancy force, N
- D particle diameter, m
- \overline{D} mean particle diameter, m
- d diameter ratio (D_1/D_2) , -
- *F* drag force, N
- \overline{F} average drag force, N
- *G* gravitational force, N
- g gravitational acceleration, m/s²
- N total hydrodynamic force, N
- *P* fluid pressure, Pa
- s density ratio, -

- u_{mf} minimum fluidization velocity, m/s
- \overline{u}_{mf} average min. fluidization vel., m/s
- V particle volume, m³
- x solids volumetric fraction, -
- y polydispersion index (Eq. 4), -

Symbols

- e voidage, -
- ϕ particle sphericity, -
- ρ solid density, kg/m³
- $\overline{\rho}$ average solid density, kg/m³

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