Chemical Engineering Communications
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713454788

RAINING OF PARTICLES FROM AN EMULSION-GAS INTERFACE IN A FLUIDIZED BED

K. S. Lim[a]; D. V. Khakhar[b]; Z. Chen[c]; P. K. Agarwal[c]
[a] Department of Chemical Engineering, University of British Columbia, Vancouver, Canada [b] Department of Chemical Engineering, Indian Institute of Technology, Bombay, India [c] Department of Chemical and Petroleum Engineering, The University of Wyoming, Laramie, WY, USA

Online publication date: 09 June 2010

To cite this Article Lim, K. S. , Khakhar, D. V. , Chen, Z. and Agarwal, P. K.(1997) 'RAINING OF PARTICLES FROM AN EMULSION-GAS INTERFACE IN A FLUIDIZED BED', Chemical Engineering Communications, 161: 1, 205 — 229
To link to this Article: DOI: 10.1080/00986449708936627
URL: http://dx.doi.org/10.1080/00986449708936627

Chem. Eng. Comm., 1997, Vol. 161, pp. 205–229
Reprints available directly from the publisher
Photocopying permitted by license only

RAINING OF PARTICLES FROM AN EMULSION-GAS INTERFACE IN A FLUIDIZED BED

K. S. LIM[b], D. V. KHAKHAR[c], Z. CHEN[a,*] and P. K. AGARWAL[a]

[a]*Department of Chemical and Petroleum Engineering, The University of Wyoming, Laramie, WY 82071, USA;*
[b]*Department of Chemical Engineering, University of British Columbia, Vancouver, Canada;*
[c]*Department of Chemical Engineering, Indian Institute of Technology, Bombay, India*

(Received 23 July 1996; In final form 28 January 1997)

This paper deals with the raining of particles from an interface between a dense fluidized phase and a gas phase with the fluidized phase uppermost. Such interfaces occur at the upper surfaces of gas bubbles and slugs in fluidized beds. Particle rain in these cases would enhance contact between gas and particles within the bubbles and slugs.

The rise velocities of single square-nosed slugs injected in incipiently fluidized beds of different diameters were measured. Relatively small columns of internal diameters of 0.0125, 0.019 and 0.0254 m were employed in the experiments; In such beds, square-nosed slugs are formed which span the entire cross-section of the beds and rise entirely due to raining of particles from their top surfaces. Since the upper surface of such slugs is flat, their motion can be analyzed using the one-dimensional hydrodynamic theory. Glass ballotini and sand of different sizes were used as bed particles. Comparison of theory and experiment has enabled the determination of the dimensionless gradient diffusivity characterizing the motion of particles induced by a gradient in the void fraction. The results confirm the scaling proposed by Batchelor (1988). The use of the calculated gradient diffusivity in the criterion for stability of a gas fluidized bed predicts the systems under consideration to be always unstable.

Keywords: Particle rain; square-nosed slug; slug rise velocity; fluidized bed; hydrodynamic model

*Corresponding author.

INTRODUCTION

A bed of particles is said to be fluidized when the drag force on the particles due to the upward flow of fluid counterbalances the gravitational and buoyant forces acting on the particles. The fluidized particles are mobile and their collective behavior, at length scales large relative to the particle size, is qualitatively similar to that of a liquid (Davidson *et al.*, 1977). The particle volume fraction in gas fluidized beds is generally not uniform at superficial gas velocities greater than the minimum fluidization velocity for Group B and D powders and the minimum bubbling velocity for Group A powders (Geldart, 1973). The excess flow traverses through the bed in the form of gas pockets (referred to as bubbles or slugs) rising through the dense fluidized phase (referred to as emulsion). The rise velocity and shape of the bubbles in the emulsion are very similar to gas bubbles rising in an inviscid liquid (Davidson *et al.*, 1977). The analogy between the emulsion-gas system and the inviscid liquid-gas system also holds for axi-symmetric and wall slugs which are observed in fluidized beds with a large length to diameter ratio. However, an important difference between the liquid-gas and emulsion-gas systems is that the emulsion-gas interface is not a true interface. In the absence of cohesive forces, the particles do not have to overcome an energy barrier to escape into the gas phase. If particles in the emulsion phase have a random (diffusive) motion about their mean position, it is possible for particles close to the emulsion-gas interface to move into the region of low particle volume fraction. The drag force on the particles that move into this region will be significantly lower than that in the emulsion phase and, consequently, the particles will fall through the lean phase to the lower surface of the bubble or slug. Thus, a continuous rain of particles from the roofs of bubbles or slugs is inevitable.

The raining of particles from the roofs of bubbles was observed early in the study of fluidized beds, and one of the first models for the rise velocity of bubbles attributed the motion of the bubbles predominantly to such particle rain (Yasui and Johanson, 1958). Later models, starting with the work of Davidson (1961), assumed that all particles flowed around the bubble, though the existence of the rain was well recognized (Jackson, 1963b). Davidson's approach provided good predictions of the bubble rise velocities and gas flow patterns around a bubble and thus has been used in a number of analyses of bubble motion (Clift and Grace, 1985). While it may be justified to neglect the particle rain in most cases in comparison with the bulk flow, the rain could affect two aspects of the system significantly: (i) particle kinematics, and (ii) gas-solids contact. In the former case,

particle rain would alter the transfer rate of the particles carried in the bubble wake to the emulsion phase, as well as the entrainment of particles due to the passage of the bubble (Gabor, 1972). In the later case, raining of particles would increase the contact between the particles and the bubble gas which is very often considered to bypass the emulsion phase with little gas-solid contact.

The case of axis-symmetric slugs and wall slugs has been treated successfully using the approach outlined above, again assuming the particle rain to be negligible (Davidson *et al.*, 1977). However, the case of square-nosed slugs falls outside the scope of the above approach. The top surface of such slugs is flat, and their rise is controlled by the raining of particles. Such slugs are formed in fluidized beds of coarse particles and large length to diameter ratios (Geldart *et al.*, 1978; Thiel and Potter, 1977). Experimental studies have shown that the rise velocities of square-nosed slugs could be lower than the other two types. However, a theoretical understanding of the system remains incomplete (Clift and Grace, 1985).

In this paper, we report measurements on the rise velocities of single square-nosed gas slugs injected in an incipiently fluidized column. Experiments have been performed in columns of different diameters. Glass ballotini and sand, of different sizes, have been used as bed particles. Data interpretation is based on the one-dimensional hydrodynamic theory; the term of particle diffusion proposed by Batchelor (1988) is also taken into account. The results of the analysis enable the determination of the gradient diffusivity characterizing the motion of particles induced by a gradient in void fraction.

EXPERIMENTAL METHODS AND RESULTS

Experiments were carried out to measure the rise velocity of a single square-nosed slug injected in an incipiently fluidized column. Such experiments have the advantage that once the slug has been injected, the height of the fluidized bed remains constant until the slug reaches the top of the bed. Mathematical analysis of such a system will be considerably easier than experiments where continuous injection of gas leads to the formation of multiple slugs in the bed and consequent periodic changes in bed height (Geldart *et al.*, 1978; Thiel and Potter, 1977). To investigate the effect of the tube diameter on the rise velocity, experiments were performed in tubes of different internal diameters, namely, 0.0125, 0.019, and 0.0254 m. Metered air from a laboratory compressor was introduced into these columns

through sintered brass distributors; the flow was controlled to maintain minimum fluidization. A separate pressurized chamber was used to feed a pulse of gas above the distributor for generating square-nosed slugs in the bed. A schematic diagram of the experimental apparatus is shown in Figure 1a. To investigate the effects of bed particle type and size, experiments were performed with glass ballotini and sand particles of different sizes. The experimental conditions are summarized in Table I. The minimum fluidization velocities of the particles were also measured and are included in this tabulation in the form of the Reynolds number at minimum fluidization, Re_{mf}. The Froude numbers based on the terminal velocity of the particles, Fr_t, were calculated using established correlations and are also presented.

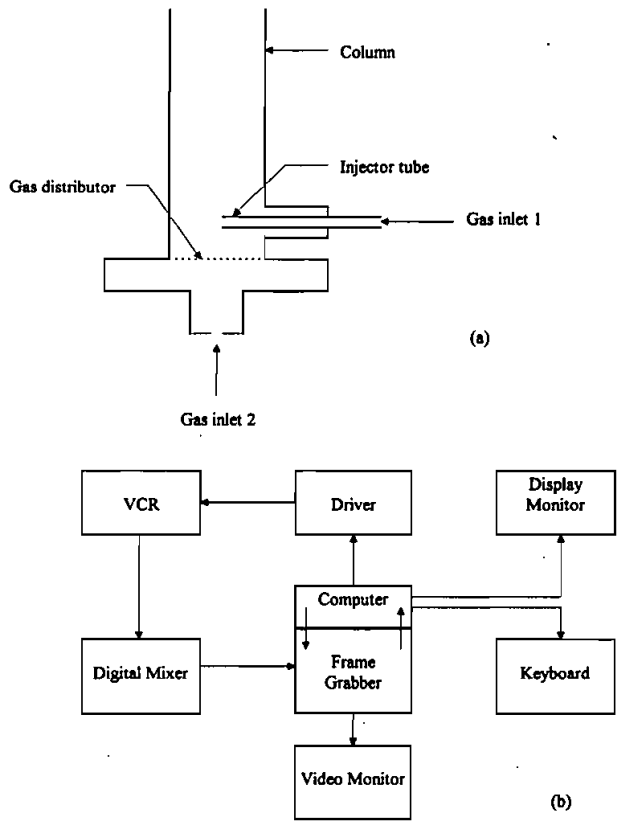

FIGURE 1 Schematic diagram of the experimental setup (a) experimental apparatus; (b) image analysis hardward configuration.

TABLE I Experimental Conditions and Results

Particles Type				*Mean slug rise velocity × 10^2, m/s column diameter, m*		
	Size × 10^3, m	Re^a_{mf}	Fr^b_t	*0.0125*	*0.0190*	*0.0254*
Glass ballotini	0.240	1.3	1137	1.73		
	0.512	7.8	2457	3.23	3.33	2.67
	0.725	17.6	3124	3.43	2.66	3.49
Sand	0.316	2.3	1573	1.85		
	0.435	4.7	2148	3.42		
	0.775	25.7	3250	4.12		4.39
	1.090	51.1	3873	4.51		4.37

[a]The minimum fluidization velocity was measured experimentally.
[b]The terminal velocity was estimated from available correlations (Fosocolo and Gibilaro, 1984; Dallavalle, 1948).

A charge-coupled device (CCD) video camera connected to a videorecorder (VCR) was used to record the rise of the slug in the bed. The measurements of the slug rise velocity were performed using a system developed in-house for the frame-by-frame analysis of image sequences from a videotape. The system comprises of a computer, VCR, and a digital video mixer and frame grabber (Fig. 1b). The key functions on the VCR, for instance, the *play*, *pause*, *frame advance* and *stop* modes, are controlled by the computer through an optocoupler driver. The digital video mixer is used to improve the signal synchonization and to eliminate image distortion, especially when the VCR is operating in the *pause* mode. The image signals from the digital video mixer are then acquired by the frame grabber and analyzed on the computer. After completing analysis on a frame, the computer automatically advances to the next frame on the tape, and the same execution routine is continued.

The boundary between the square-nosed gas slug and the emulsion phase is sharp and was located readily using global threshholding of the digitized image (Lim *et al.*, 1990). The measured locations of the boundary were plotted as a function of time and a linear regression of the data enabled the determination of the average slug velocity; a typical example is shown in Figure 2. From the data, the instantaneous slug velocity was also determined as a function of time. The instantaneous velocity measurements indicated that the slugs, in general, have a somewhat jerky upward motion. In some cases, slow square-nosed slugs were found to accelerate in a very short time to velocities close to the expected rise velocity of an axi-symmetric slug. Such slugs could either travel through the column at the higher velocity or decelerate, again in a very short time, to the lower velocity typical of

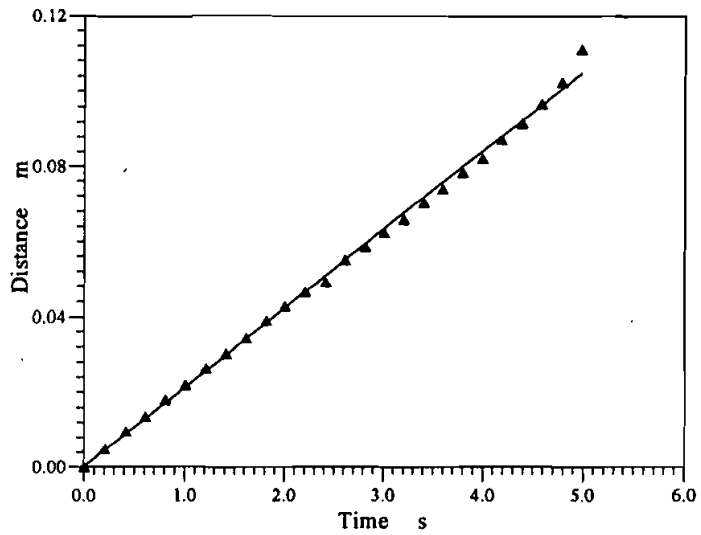

FIGURE 2 Progression of the slug interface as a function of time: Glass ballotini, $d_p = 0.512 \times 10^{-3}$ m, $D_t = 0.0254$ m, Slope $= u_{st} = 0.021$ m/s.

the square-nosed slug. An example of the rapid acceleration followed by deceleration is given in Figure 3. For the smaller particle sizes, the slugs were found at times to stop completely, forming a solid bridge across the column. A small perturbation, either manual or from alteration of the gas flow through the distributor, was sufficient to get the slug moving through the bed again. When these solid bridges formed, the solid region directly above the interface appeared to be defluidized. At the position of a small distance above the interface, however, the bed appeared to be fluidized and occasional bubbles were observed. This observation is similar to that reported by Thiel and Pottor (1977), who noted that the solid bridges behaved as a distributor for the gas flow. It should be stressed that when slugs moved steadily, the region above the slug was fluidized.

Only slugs showing relatively steady rise through the bed were selected for futher data analysis. The mean slug rise velocity is plotted as a function of bed particle size in Figure 4. The data on the measured slug rise velocities for different bed particle types and sizes as well as fluidized column diameters are given in Table I. Each measurement of the slug rise velocity represents a mean of at least thirty measurements; the actual data were spread about $\pm 25\%$ from this mean value.

The experimental results establish that the slug rise velocity depends on particle diameter. The results also indicate that, for the experimental condi-

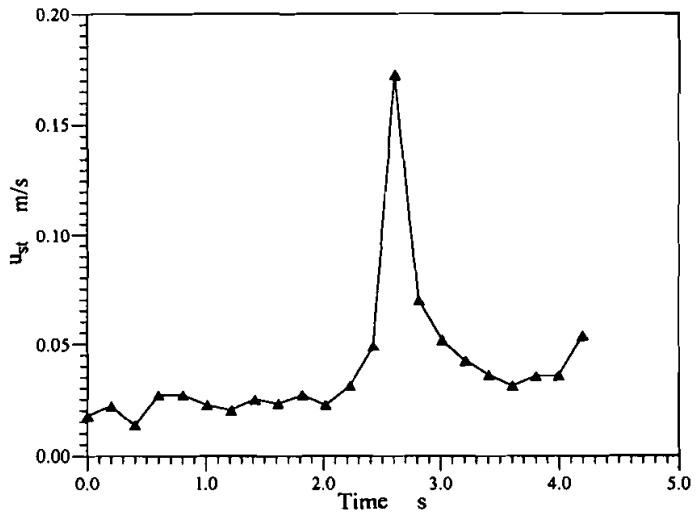

FIGURE 3 Instantaneous slug rise velocity showing rapid acceleration followed by deceleration of the slug: Glass ballotini, $d_p = 0.512 \times 10^{-3}$ m, $D_t = 0.0254$ m.

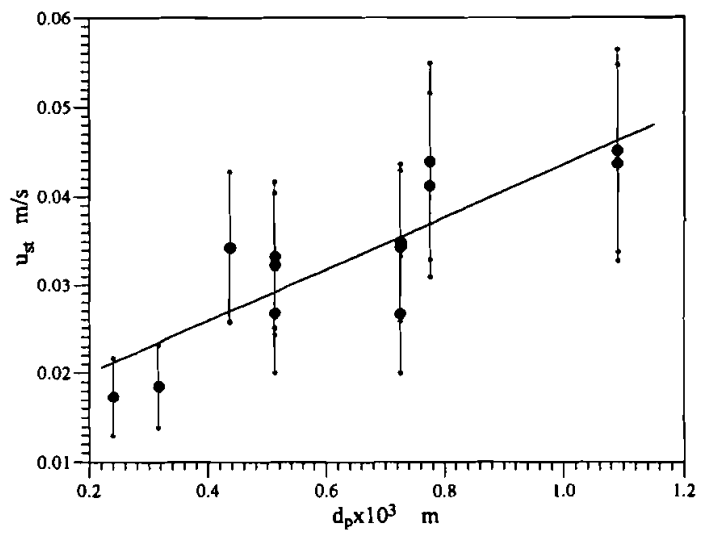

FIGURE 4 Slug rise velocity as a function of particle diameter: experimental results.

tions considered, the slug velocity appears to be independent of the tube diameter. The range of tube diameters considered in the experimental program is comparatively small (0.0125 to 0.0254 m). Additional experiments

were attempted in a larger column of 0.05 m internal diameter. However, single slugs were found to degenerate rapidly to axi-symmetric and/or wall slugs. The factors controlling the formation of square-nosed slugs have not been clearly understood. Thiel and Potter (1977) suggested that the tendency for the formation of such slugs increases with (i) the decrease in the angle of the internal friction of particles; and (ii) the decrease in the tube diameter. Baker and Geldart (1978), however, found experimental evidence contradicting the first observation and suggested that tube diameter plays a more significant role. More recently, Chen *et al.* (1997) reported that particle density also has a remarkable effect on the formation of square-nosed slugs. They found that for particles having a density similar to glass ballotini, square-nosed slugs occur only if the ratio of particle to tube diameters is higher than about 0.006. Experiments on the larger diameter columns also indicated that, though single slugs degenerate, continuous injection of gas leads to apparently more stable square-nosed slugs which, at times, coexist with wall and/or axi-symmetric slugs. Visual inspection, however, did indicate that for low flow rates in excess of that required for minimum fluidization, the rise velocities of square-nosed slugs could not have been very different from the measurements in a 0.0125 m diameter tube.

It is evident that the square-nosed slug rise velocity is substantially lower than the expected velocities for axi-symmetric or wall slugs for the glass ballotini and sand particles used in our experiments. Though Baker and Geldart (1978) found that for PVC beads the rise velocity of the square-nosed slug was of the same magnitude as axi-symmetric or wall slugs, our experimental results suggest that the square-nosed slug velocity cannot be taken to be proportional to the square root of tube diameter as reported by Chen and Fan (1988).

THEORY

Governing Equations for Steady One-Dimensional Flow

The governing equations for the flow of particles and fluid in a fluidized bed have been the subject of several investigations starting with the initial studies of Jackson (1963a) and Pigford and Baron (1965). While a definitive theory for the flow in a fluidized bed does not exist as yet, considerable progress has been made in identifying the important physical processes. The work to date, in the context of the stability of a uniformly fluidized bed has been reviewed by Jackson (1985), Batchelor (1988) and Ham *et al.* (1990).

The previous studies can be classified into two categories based on the approaches taken: (i) two-fluid equations (for example, Anderson and Jackson 1967; Homsy *et al.*, 1980); (ii) averaged equations (for example, Drew, 1980). In the first approach, the particles and fluid are taken to be interpenetrating continua and the rheological properties and the interactions between the continua are either postulated or obtained empirically. In the second approach, the microscopic equations for the emulsion are averaged over the configurations of the particles or over a material volume to obtain continuum equations for the solids and fluid flows. The averaged equations, in general, are not complete and require additional closure equations for the higher order averages. In the following, we use the recently proposed one-dimensional theory of Batchelor (1988) which nominally follows the second approach described above; however, physical arguments are used rather than a rigorous averaging procedure.

Consider the square-nosed slug to be represented by a flat dense-lean interface moving at a steady velocity, u_{st}. Neglecting radial variations, a one-dimensional description will be adopted. To derive the appropriate governing equations, the frame of reference is fixed at the interface and far from the interface the particles are assumed to be uniformly fluidized. Further, gas and particles flows are assumed to be positive in the upward direction. The average particle velocity with respect to the interface is denoted as v; the interstitial gas velocity with respect to the interface is denoted as u. Then the continuity equations for the flow of the particles and gas are

$$\frac{d(\phi v)}{dx} = 0 \quad \text{or} \quad \phi v = n = \text{constant} \tag{1}$$

$$\frac{d[(1-\phi)u]}{dx} = 0 \quad \text{or} \quad (1-\phi)\,u = q = \text{constant} \tag{2}$$

respectively, where n is the total flux of the particles and q is the total flux of the gas with respect to the moving frame of reference where $x = 0$ represents the interface location at any given time.

When the inertial terms pertaining to the gas flow are neglected, the momentum balance equations for the flow of particles and gas are

$$\rho_p \phi v \frac{dv}{dx} = -\rho_p \phi g + F_b + F - E\frac{d\phi}{dx} + \frac{d}{dx}\left(\phi \rho_p \eta \frac{dv}{dx}\right) \tag{3}$$

$$\frac{dP}{dx} = -F \tag{4}$$

where E is the elasticity modulus of the emulsion phase. F_b, the buoyancy force has been the subject of intense debate recently: some investigators suggest that it should be based on the fluid density alone (Epstein, 1984; Clift *et al.*, 1987; Jean and Fan, 1992); others base it on the density of the gas-particle suspension (Barnea and Mizrahi, 1973; Foscolo and Gibilaro, 1984). Consistent with the expression for the drag force employed below, we adopt $F_b = \phi\rho_b g$, in which ρ_b is the bed density, $\rho_b = \phi\rho_p + (1-\phi)\rho_f \approx \phi\rho_p$. It is believed that the application of different expressions for the buoyancy would not affect the conclusion drawn in this paper provided that the corresponding correlation for the drag force is used. The gas momentum balance specifies the gas pressure profile, which can be obtained independently given the variation of the hydrodynamic force. The fourth term on the right hand side of Equation 3 represents a force induced by the presence of a void fraction gradient. Such a term has been included in several previous analyses (Anderson and Jackson, 1967; Wallis, 1962, 1969; Foscolo and Gibilaro, 1984); the novel feature of Batchelor's approach is to relate the origin of this term to the diffusion of particles.

The random diffusive motion of particles in suspensions occurs due to multiparticle interactions and has become the focus of study only recenlty. The case of interest here is the diffusion of particles in the presence of a solids volume fraction gradient. Two other cases have received attention: self-diffusion resulting from shearing of a suspension (Leighton and Acrivos, 1987); and self-diffusion of particles in a statistically homogenous sedimenting suspension (Ham and Homsy, 1988). For our case, the gradient in the particle volume fraction results in a particle diffusive flux given by

$$j = -D\frac{d\phi}{dx} \tag{5}$$

where D is the gradient diffusivity. Note that j is the flux with respect to the moving frame of reference. The particle velocity appearing in Equation 1 and 3 is based on the total flux of particles, n, which represents the sum of convective and diffusive fluxes. It should be noted that the interface is defined as the horizontal surface at which the diffusional flux is maximum.

Specification of the material functions – E and η – and the hydrodynamic force, F, is required to complete the governing equations. For our analysis, it is assumed that the contribution of emulsion phase viscous transfer can be neglected. It is important while making this assumption to consider

whether models which neglect particle phase viscosity can lead to solutions which resemble slugs observed in the narrow diameter columns. Earlier non-linear analyses reported by Needham and Merkin (1983) and Fanucci *et al.* (1981) indicated that inclusion of particle phase viscosity, even small, was necessary to obtain slug-like solutions. More recently, however, Ganser and Lightbourne (1991) have shown analytically that oscillatory traveling wave solutions resembling slugs are obtained even when particle phase viscosity term is neglected. Their results were summarized in the form of a theorem for the existence of slug-like solutions; it can be easily verified that the model considered in this paper satisfies the requirements of their theorem.

The hydrodynamic interaction force is taken to be

$$F = \frac{\rho_p g}{u_t^m} |u_s|^{m-1} u_s (1-\phi)^p \phi \tag{6}$$

where $u_s = (1-\phi)(u-v)$ is the superficial slip velocity between particles and the fluid. In Equation 6, m is the drag-slope parameter which depends on the particle Reynolds number and is related to the Richardson-Zaki index, n_i, as $m = 4.8/n_i$. A common voidage dependency (Foscolo *et al.*, 1983) has been chosen for the different flow regimes, and the value of p is taken to be -3.8. For slow flow conditions Equation 6 becomes identical to that used by Needham and Merkin (1983).

The effective elasticity of the particles can be considered to be made up of two contributions: the first arising from the mean square velocity fluctuations in a homogeneous bed; and the second due to hydrodynamic dispersion down a concentration gradient (Batchelor, 1988; Ham *et al.*, 1990). Batchelor suggests that the first contribution is relatively small. The experimental results of Ham *et al.* (1990) do suggest otherwise; however, for the present analysis it is assumed that

$$E(\phi) = \frac{D}{BV_p}. \tag{7}$$

The validity of this assumption will be evaluated from interpretation of our experiments in a later section. Batchelor (1988) defined the bulk mobility; B, as "ratio of the (small additional) mean particle velocity relative to zero-volume flux axes, to the (small additional) steady force applied to each particle of a homogeneous dispersion". Consequently, when the gas

and particle fluxes are given by Equations 1 and 2,

$$B = \left[\frac{\partial F_p}{\partial u_s}\right]^{-1} = \left[\frac{V_p}{\phi}\frac{\partial F}{\partial u_s}\right]^{-1} \tag{8}$$

where F_p is the drag force on an individual particle in the suspension and is related to the total drag on particles according to $F_p = V_p F/\phi$. To specify diffusivity, D, following Batchelor (1988) we use

$$D = \frac{\alpha d_p}{2}|u_s| \tag{9}$$

where the dimensionless gradient diffusivity, α, is assumed to be a constant.

With the assumptions described above, and using the continuity equations, the governing momentum balance (Equation 3) for the particles can be rewritten as

$$\left[\frac{n^2}{g\phi^2} - \frac{m\alpha d_p}{2u_t^m}(1-\phi)^{p+m}(u-v)^m\right]\frac{d\phi}{dx} = \phi(1-\phi) - \left(\frac{u-v}{u_t}\right)^m (1-\phi)^{p+m}\phi. \tag{10}$$

The boundary condition for the particle volume fraction far from the interface is

$$\phi = \phi_0 = \text{constant}. \tag{11}$$

The corresponding gas and solids velocities are related by

$$\phi_0 g(\rho_p - \rho_b) = F(\phi_0, u_0, v_0) \tag{12}$$

which is obtained from Equation 10 using the boundary condition, Equation 11. The continuity equations yield

$$v\phi = n = v_0\phi_0 = \text{constant} \tag{13}$$

$$(1-\phi)u = q = (1-\phi_0)u_0 = \text{constant} \tag{14}$$

where v_0 and u_0 are, respectively, the average particle and gas linear velocity (in the moving frame of reference) far from the interface.

It is clear that the diffusional flux vanishes far from the interface, and by definition it is maximum at the interface; thus

$$\frac{dj}{dx} = 0 \qquad x = 0. \tag{15}$$

Also, to satisfy continuity at the interface, the particle velocity must be the same as the diffusional velocity

$$\frac{n}{\phi} = -\frac{D}{\phi}\frac{d\phi}{dx} \qquad x = 0 \tag{16a}$$

which can be rewritten as

$$n = j = -D\frac{d\phi}{dx} \qquad x = 0. \tag{16b}$$

The raining of particles from the interface is, thus, a consequence of the diffusion of particles across the gradient in void fraction. It is worth noting that the bulk flow term does not appear in these equations since they are based on a frame of reference which moves upwards with the slug.

Defining dimensionless fluxes $N = n/u_t$, $J = j/u_t$ and $Q = q/u_t$, and the dimensionless distance as $X = x/d_p$, Equations 10, 12, 15 and 16 can then be written in a dimensionless form as

$$\left[\frac{Fr_t N^2}{\phi^2} - \frac{m\alpha}{2}(1-\phi)^p\left|N + Q - \frac{N}{\phi}\right|^m\right]\frac{d\phi}{dX} = \phi(1-\phi) - \left|N + Q - \frac{N}{\phi}\right|^m (1-\phi)^p\phi \tag{17}$$

$$\left|N + Q - \frac{N}{\phi_0}\right|^m (1-\phi_0)^{p-1} = 1 \tag{18}$$

$$N = J \qquad X = 0 \tag{19}$$

$$\frac{dJ}{dX} = 0 \qquad X = 0 \tag{20}$$

where $Fr_t = u_t^2/gd_p$. Equations 17–20 need to be solved to characterize the consequences of the diffusive motion of particles at the interface.

Solution Procedure and Results

The calculation of the unknown parameters N, Q and the dimensionless gradient diffusivity, α, was decoupled from the computation of the solids volume fraction profile by modifying the boundary conditions at the interface (Equations 19 and 20) to

$$N = J \qquad \phi = \phi_i \tag{21}$$

$$\frac{dJ}{d\phi} = 0 \qquad \phi = \phi_i \tag{22}$$

where the diffusive flux, obtained from Equations 5 and 17, is

$$J = \frac{\alpha\phi\left|N + Q - \frac{N}{\phi}\right|\left\{\left|N + Q - \frac{N}{\phi}\right|^m (1-\phi)^p - (1-\phi\right\}}{\frac{2Fr_t N^2}{\phi^2} - m\alpha(1-\phi)^p\left|N + Q - \frac{N}{\phi}\right|^m} \tag{23}$$

N, Q and α were obtained by the simultaneous solution of the non-linear algebraic Equations 18, and 21 and 22 using specified values of $\phi_0 = 0.58$ and $\phi_i = k\phi_0$, where k is a constant. As discussed later, $k = 0.83$ leads to good agreement between measured and predicted slug rise velocities. The volume fraction profile was then obtained by numerical integration of Equation 17 for positive and negative X, using $\phi = \phi_i$ at $X = 0$.

Figure 5 shows the particle volume fraction profiles for three different particles. It can be seen that equilibrium is reached within one particle diameter above the interface. Below the interface, the solid volume fraction decays rapidly in about 4 to 8 particle diameters, essentially depending on the particle size: the larger the particle size is, the longer will be the distance to reach the equilibrium; then it approaches gently the unity gas voidage. The diffusional flux profiles, also plotted as a function of X in Figure 6, show a distinct maximum at the interface and the fluxes approach zero at the two sides of the interface. It can be found that the flux at the interface increases with the increase in the particle diameter: this is in agreement with the correlation proposed by Wallis (1969), which shows that the drift flux is proportional to the terminal velocity of particles.

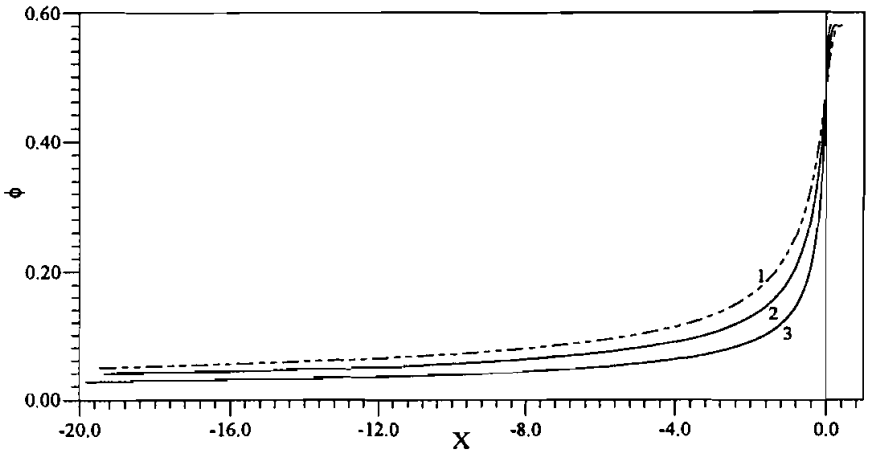

FIGURE 5 Solid volume fraction as a function of dimensionless distance $d_p \times 10^3$(m):(1) 0.775; (2) 0.512; (3) 0.316.

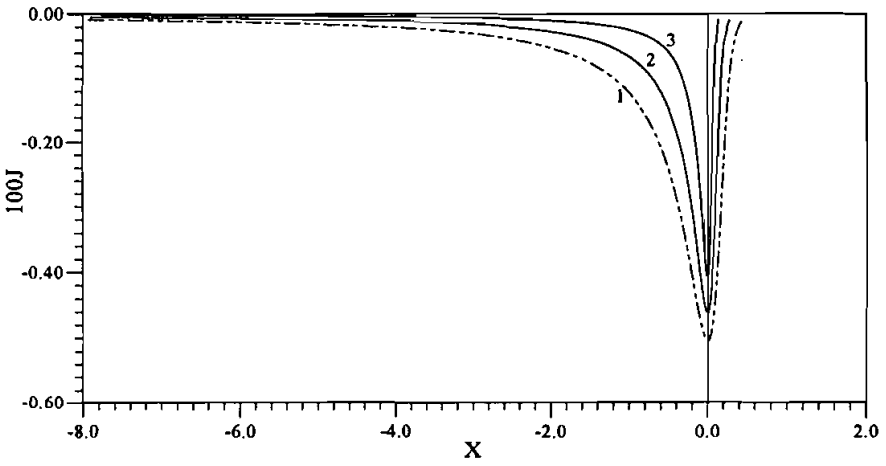

FIGURE 6 Dimensionless diffusion flux as a function of dimensionless distance $d_p \times 10^3$(m):(1) 0.775; (2) 0.512; (3) 0.316.

DISCUSSION

During the rise of a single slug in an incipiently fluidized bed, the height of the bed remains constant and the particle velocity with respect to the container wall far from the interface is zero. Consequently, the total particle

flux, n, in the moving frame of reference is given by

$$n = -\phi_0 u_{st}. \tag{24}$$

Thus, the rise velocity of slugs can be determined from the hydrodynamic model described above. Figure 7 compares model calculations with experimental data on slug rise velocities using $k = 0.83$; reasonable agreement is obtained. The calculated values of the dimensionless gradient diffusivity, α, are shown in Figure 8 as a function of particle diameter. It appears, in agreement with the theory of Batchelor (1988), that α increases with the increase of particle size.

An alternate interpretation of data is possible if the slug rise is represented as the motion of a dynamic wave propagating through the fluidized column (Wallis, 1962, 1969). The dynamic wave velocity, u_d, can be shown to be

$$u_d = \pm\left(\frac{E}{\rho_p}\right)^{0.5}\bigg|_{\phi=\phi_0} = \pm u_t\left[\frac{m\alpha(1-\phi_0)}{2Fr_t}\right]^{0.5}. \tag{25}$$

With u_d taken to be the slug rise velocity, Equation 25 can be used to calculate α as well. The values of α so calculated are compared with the present model in Figure 9. It can be seen that Equation 25 overestimates the calculated dimensionless gradient diffusivity; perhaps α, contrary to the

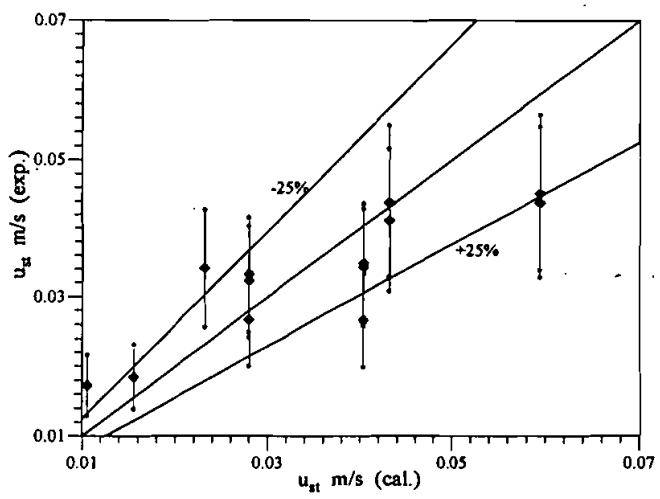

FIGURE 7 Comparison of measured and predicted slug rise velocities.

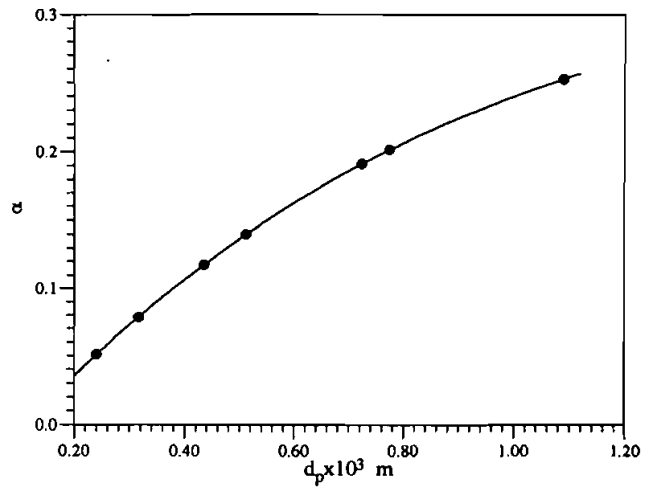

FIGURE 8 Dimensionless gradient diffusivity vs particle diameter.

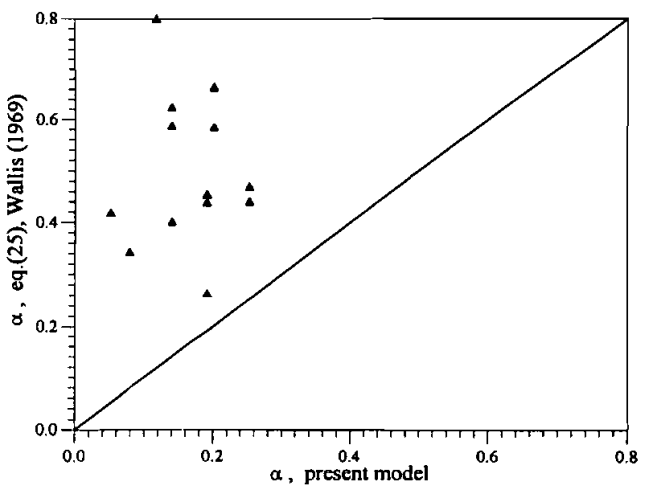

FIGURE 9 Comparison of calculated dimensionless gradient diffusivity and Walli's theory.

assumption in our analysis, varies with the solid volume fraction. Nevertheless, the magnitudes of α obtained from the two approaches are the same.

Comparison with Previous Data

Very few measurements of the hydrodynamic diffusivity have been reported in the literature. Davis and Hassen (1988, 1989) have reported data on the

spreading of an interface in dilute ($0.002 < \phi_0 < 0.15$) sedimenting suspensions of small particles in a liquid. The observed spreading in excess of that expected from polydispersity and hindered settling effects could be described approximately by a diffusion process. Ham and Homsy (1988) observed the motion of an individual sphere settling in the midst of a suspension of the spheres for $0.025 < \phi_0 < 0.1$ under slow flow conditions. Fluctuations in settling speed were interpreted in terms of a diffusion process. The values for the dimensionless self-diffusion coefficient were very similar to the measurements reported by Davis and Hassen (1989). It should be mentioned that self-diffusion also scales, similar to gradient diffusion, with characteristic speed and particle size; however, the dimensionless diffusivity so obtained has been found to be a function of the Reynolds number reflecting its inertial origin. The data discussed above, in terms of the dimensionless hydrodynamic diffusivity, α (scaled with respect to particle radius and reflecting self-diffusion and/or gradient diffusion), are plotted as a function of ϕ_0 in Figure 10.

More recently Ham *et al.* (1990) have measured the voidage and velocities at minimum fluidization and at critical conditions for the onset of the instability waves in liquid fluidized beds. A wide range of particle and fluid properties was examined. Using the results of linear stability theory for data interpretation, they concluded that the elasticity was a function of the Reynolds number and, hence, was of inertial origin. The dimensionless

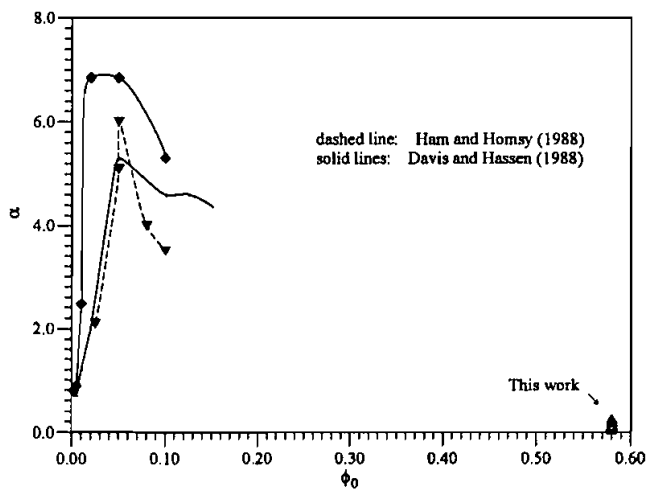

FIGURE 10 Dimensionless hydrodynamic diffusivity as a function of the bulk particle volume fraction.

gradient diffusivity parameter was deduced to be very small: 2.3×10^{-3}. The reason for the discrepancy between these measurements and ours is unclear.

Wallis (1962) has reported bed-support experiments in which the upward flowing fluid supported a packed layer of particles against a grid. Reduction in fluid velocity led to particles falling off from the packed layer. The velocity of progression of the interface, u_{st}, was measured as a function of the fluid velocity and the results were interpreted in terms of the propagation of a dynamic wave. Extrapolation of his results to the situation where the fluid velocity equals the minimum fluidization velocity leads to $u_{st}(gd_p)^{-1/2} \approx 0.5$–$2$, whereas our experimental data yield $u_{st}(gd_p)^{-1/2} \approx$ 0.3–0.6. with an average of 0.42. These results are not incompatible and the difference is thought to arise from an important feature which distinguishes our experimental technique from that used by Wallis: the layer of particles supported against the grid will undoubtedly be in close packed condition whereas in our experiments the layer above the slug is fluidized.

Stability of a Gas-Fluidized Bed

According to Wallis (1962, 1969), the criterion for instability is that the kinematic wave speed exceeds the dynamic wave speed. Batchelor (1988) has shown this to be correct if acceleration and reaction terms in the momentum balance equation are neglected.

The kinematic wave speed can be evaluated from the correlation of Slis *et al.* (1961)

$$u_k = \phi \left.\frac{du_s}{d(1-\phi)}\right|_{\phi=\phi_0} \tag{26}$$

Under equilibrium conditions (the drag force on a particle counterbalances the net weight of the particle), it can be shown that the superficial slip velocity, u_s, is given by

$$u_s = (1-\phi)^{(1-p)/m} u_t. \tag{27}$$

Substituting Equation 27 into Equation 26 yields

$$u_k = u_t\left(\frac{1-p}{m}\right)\phi_0(1-\phi_0)^{(1-p-m)/m}. \tag{28}$$

The critical condition for stability can thus be obtained by combining Equations 25 and 28 as

$$\sqrt{\frac{m\,\alpha(1-\phi_0)}{2Fr_t}} = \left(\frac{1-p}{m}\right)\phi_0(1-\phi_0)^{(1-p-m)/m}. \tag{29}$$

For $m = 1$, Equation 29 reduces to the forms proposed by Batchelor (1988) and Ganser and Lightbourne (1991).

Using the calculated value of α, prediction of the data on voidage at minimum bubbling for glass ballotini and sand particles was attempted; the results show that fluidized beds made of the particles used in this work are always unstable in a range of $0 \leqslant \phi_0 \leqslant 0.6$; Calculations were also performed to compare Equation 29 with the criterion for the stability of fluidized beds developed by Foscolo and Gibilaro (1984), which has been shown to predict quantitatively the minimum bubbling voidage in good agreement with various experimental observations (Foscolo and Gibilaro, 1987). $(u_k - u_d)/u_t$ calculated using the two approaches are compared in Figure 11; the agreement is excellent.

Concluding Remarks

The experimental results on the rise velocity indicate about 25% variation about the mean. This variation could arise from the size distribution of the fractions used as bed particles in the experiments. Clearly, further research must employ narrower size distributions. Sand and glass ballotini have roughly the same particle density (≈ 2500 kg/m^3). The results for these particles are comparable over the entire range of particle sizes considered in our experiments. Further research will be necessary to investigate the effect of particle density on the slug rise velocity, especially in the light of the results for PVC particles obtained by Baker and Geldart (1978) and the results reported by Chen *et al.* (1997), which show that the slug type also depends on the particle density.

Another aspect which requires further discussion is the effect of wall friction on the slug rise velocity; the results reported in the literature are somewhat contradictory. Jones and Leung (1985) note that for a fluidized section with solids volume fraction close to that corresponding to minimum fluidization, the effect of particle-wall friction can be safely neglected. Baker and Geldart (1978), in their investigation on slugging in beds of coarse particles, found that the pressure drop across the bed exceeded the weight of

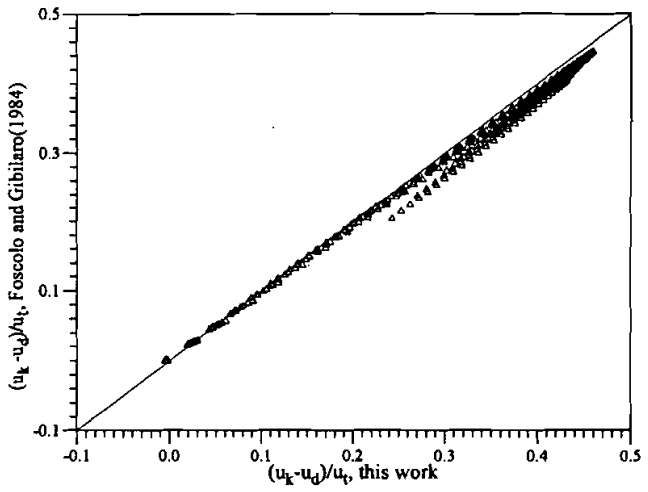

FIGURE 11 Comparison of the criteria of this work and of Foscolo and Gibilaro (1984): all particles used in this work; $0 \leqslant \phi_0 \leqslant 0.60$.

the solids in the bed. This excess pressure loss was attributed to frictional losses. It should be noted, however, that for PVC beads – the only type of solids exhibiting square-nosed slugging – this excess pressure loss was very small. The presence of friction losses would imply that the slug rise velocity should depend on tube diameter. The absence of such dependency in our experimental results, then, indicates that wall friction losses are not important. However, it remains difficult to explain the solids bridging observed in some runs. Stewart and Davidson (1967) suggested that a region of positive particle pressure near the wall and just below the nose of an axi-symmetric slug can lead to the locking of particles to form a defluidized zone and particle bridges. When a stationary solid bridge forms above a square-nosed slug, as noted earlier, the existence of a defluidized region separating the gas-emulsion boundary and a fluidized region can be observed visually. How and why this defluidized region develops is not clear and should require further investigation.

CONCLUSIONS

Diffusion of particles induced by a void-fraction gradient will inevitably lead to raining of particles from the top surface of a bubble or a slug in a gas fluidized bed. This paper considers the motion of a flat gas-emulsion

interface in an incipiently fluidized bed. Experiments were conducted to measure the rise velocities of single injected square-nosed slugs in columns maintained at minimum fluidized conditions. The top surface of such slugs is flat, and they rise entirely by the raining of particles from the particulate phase. Tubes of different diameters and bed particles (sand and glass ballotini) of different sizes were employed. The rise velocities of the square-nosed slugs were found to depend on the bed particle size and to be independent of the tube diameter. The rise velocities were also found to be substantially lower than those expected for wall or axi-symmetric slugs. The experimental data were analyzed using a one-dimensional hydrodynamic model. The results confirm the scaling for gradient diffusion proposed by Batchelor (1988) – the dimensionless diffusivity was found to increase with the increase of the particle diameter. The calculated value of gradient diffusivity used in the criterion for stability of gas fluidized beds is able to predict that the systems under study are always unstable as expected in the whole range of possible voidage fractions.

Acknowledgements

The experimental work was conducted at the University of Adelaide with financial support from the Australian Research Council. The authors would like to thank K. Brown and T. Jones for assistance with the experimental work and K. Kelly for assistance in preparing the manuscript.

NOMENCLAUTURE

B	bulk mobility, defined in Equation 8
d_p	particle diameter, m
D	gradient diffusivity, m^2/s
D_t	column diameter, m
E	elasticity modulus of emulsion phase, N/m^2
F	hydrodynamic drag force per unit volume, N/m^3
F_b	buoyancy force per unit volume, N/m^3
F_p	drag force on a single particle, N
Fr_t	Froude number based on the terminal velocity of a particle
g	acceleration due to gravity, m/s^2
j	diffusive flux of particles, m/s
J	dimensionless diffusive flux of particles

k	constant
m	drag slope parameter
n	total flux of particles with respect to the moving frame or reference, m/s
n_i	Richardson-Zaki index
N	dimensionless flux of particles
P	gas pressure, kPa
q	total gas flux with respect to the moving frame of reference, m/s
Q	dimensionless gas flux
Re_{mf}	Reynolds number at minimum fluidization
u	interstitial gas velocity with respect to the moving interface, m/s
u_d	dynamic wave velocity, m/s
u_k	kinematic wave velocity, m/s
u_s	superficial slip velocity between particles and fluid, m/s
u_{st}	rise velocity of a slug, m/s
u_t	terminal velocity of particles, m/s
v	particle velocity with respect to the moving interface, m/s
V_p	particle volume, m^3
x	axial distance from the moving interface, m
X	dimensionless axial distance from the interface

Greek symbols

α	dimensionless gradient diffusivity, Equation 9
η	bed viscosity, m^2/s
ρ_p	particle density, kg/m^3
ϕ	particle volume fraction

Subscripts

0	far from the interface
i	at the interface

References

Anderson, T. B. and Jackson, R. (1967) A fluid mechanical description of fluidized beds: equations of motion, *Ind. Engng Chem. Fundam.*, **6**, 527–537.

Baker, C. G. J. and Geldart, D. (1978) An investigation into slugging characteristics of large particles, *Powder Technol.*, **19**, 177–187.

Barnea, E. and Mizrahi, J. (1973) A generalized approach to the fluid dynamic of particulate systems, *Chem. Engng. J.*, **5**, 171–189.

Batchelor, G. K. (1988) A new theory for the instability of a uniform fluidized bed, *J. Fluid Mesh.*, **193**, 75–110.

Chen, Y.-M. and Fan, L.-S. (1988) Vertical slug flows of gas-solid transport in circulating fluidized beds; *Circulating Fluidized Bed Technology II* (Eds. Basu, P. and Large, J. F.) Pergamon Press, New York, 65–71.

Chen, Z., Gibilaro, L. G. and Foscolo, P. U. (1997) Fluid pressure loss in slugging fluidized beds, *Chem. Engng. Sci.*, **52**, 55–62.

Clift, R. and Grace, J. R. (1985) Continuous bubbling and slugging, in *Fluidization* (Ed. Davidson, J. F., Clift, R. and Harrison, D.), Academic Press, New York 73–132.

Clift, R., Seville, J. P. K., Moore, S. C. and Chavarie, C. (1987) Comments on buoyancy in fluidized beds, *Chem. Engng. Sci.*, **42**, 191–194.

Dallavalle, J. M. (1948) *Micromeritics*, Pitman Publishing Corporation, New York.

Davidson, J. F. (1961) Symposium on fluidization, discussion, *Trans. Inst. Chem. Engrs.*, **39**, 230–232.

Davidson, J. F., Harrison, D. and Guedes de Carvalho, J. R. (1977) On the liquid-like behavior of fluidized beds, *Ann. Rev. Fluid Mech.*, **9**, 55–86.

Davis, R. H. and Hassen, M. A. (1988, 1989) Spreading of the interface at the top of a slightly polydisperse sedimenting suspension, *J. Fluid Mesh.*, **196**, 107–134; Corrigendum: *J. Fluid Mech.*, **202**, 598–599.

Drew, D. A. (1980) Mathematical modeling of two phase flow, *Ann. Rev. Fluid Mesh.*, **15**, 261–291.

Epstein, N. (1984) Comments on a unified model for particulate expansion of fluidized beds and flow in fixed porous media, *Chem. Engng. Sci.*, **39**, 1533–1534.

Fanucci, J. B., Ness, N. and Yen, R.-H. (1981) Structure of shock waves in gas-particle fluidized beds, *Phys. Fluids*, **24**, 1944–1954.

Foscolo, P. U., Gibilaro, L. G. and Waldram, S. P. (1983) A unified model for particulate expansion of fluidized beds and flow in fixed porous media, *Chem. Engng. Sci.*, **38**, 1251–1260.

Foscolo, P. U. and Gibilaro, L. G. (1984) A fully predictive criterion for the transition between particulate and aggregative fluidization, *Chem. Engng. Sci.*, **39**, 1667–1675.

Foscolo, P. U. and Gibilaro, L. G. (1987) Fluid dynamic stability of fluidized suspensions: the particle bed model, *Chem. Engng. Sci.*, **42**, 1489–1500.

Gabor, J. D. (1972) On the mechanics of fluidized particle movement, *Chem. Engng. J.*, **4**, 118–126.

Ganser, G. H. and Lightbourne, J. H. (1991) Oscillatory traveling waves in a hyperbolic model of a fluidized bed, *Chem. Engng. Sci.*, **46**, 1339–1347.

Geldart, D. (1973) Types of gas fluidization, *Powder Technol.*, **7**, 285–295.

Geldart, D., Hurt, J. M. and Wedia, P. H. (1978) Slugging in beds of large particles, *AIChE Symp. Ser.*, **176**, 74, 60–66.

Ham, J. M. and Homsy, G. M. (1988) Hindered settling and hydrodynamic dispersion in quiescent sedimenting suspensions, *Int. J. Multiphase Flow*, **14**, 533–546.

Ham, J. M. Thomas, S., Guazzelli, E., Homsy, G. M. and Anselmet, M. C. (1990) An experimental study of the stability of liquid-fluidized beds, *Int. J. Multiphase Flow*, **16**, 171–185.

Homsy, G. M., El-Kaissy, M. M. and Didwania, A. (1980) Instability waves and the origin of bubbles in fluidized beds–II, *Int. J. Multiphase Flow*, **6**, 305–318.

Jackson, R. (1963a) The mechanics of fluidized beds, I. The stability of the uniform state of fluidization, *Trans. Inst. Chem. Engrs.*, **41**, 13–21.

Jackson, R. (1963b) The mechanics of fluidized beds, II. The motion of fully developed bubbles, *Trans. Inst. Chem. Engrs.*, **41**, 22–28.

Jackson, R. (1985) Hydrodynamic stability of fluid-particle systems in *Fluidization* (Eds. Davidson, J. F., Clift, R. and Harrison, D.), Academic Press, New York, 47–72.

Jean, R.-H. and Fan, L.-S. (1992) On the model equations of Gibilaro and Foscolo with corrected buoyancy force, *Powder Technol.*, **72**, 201–206.

Jones, P. L. and Leung, L. S. (1985) Downflow of solids through pipes and valves in *Fluidization* (Eds. Davidson, J. F., Clift, R. and Harrison, D.), Academic Press, New York, 293–329.

Leighton, D. and Acrivos, A. (1987) Measurement of shear induced coefficient of self-diffusion in concentrated suspensions of solid spheres, *J. Fluid Mech.*, **177**, 109–131.

Lim, K. S., O'Neill, B. K. and Agarwal, P. K. (1990) Measurement and modeling of bubble parameters in a two-dimensional gas-fluidized bed using image analysis, *Powder Technol.*, **60**, 159–171.

Needham, D. J. and Merkin, J. H. (1983) The propagation of a voidage disturbance in a uniformly fluidized bed, *J. Fluid Mech.*, **131**, 427–454

Pigford, R. L. and Baron, T. (1965) Hydrodynamic stability of a fluidized bed, *Ind. Engng. Chem. Fundam.*, **1**, 81–87.

Richardson, J. F. and Zaki, W. N. (1954) Sedimentation and fluidization: Part 1, *Trans. Inst. Chem. Engrs.*, **32**, 35–52.

Slis, P. L., Willemse, Th. W. and Kramers, H. (1959) Response of the level of a liquid fluidized bed to a sudden change in the fluidizing velocity, *Appl. Sci. Res.*, **A 8**, 209–218.

Stewart, P. S. B. and Davidson, J. F. (1967) Slug flow in fluidized beds, *Powder Technol.*, **1**, 61–80.

Thiel, W. J. and Potter, O. E. (1977) Slugging in fluidized beds, *Ind. Engng Chem. Fundam.*, **16**, 242–247.

Wallis, G. B. (1962) *One-dimensional waves in two-component flow*, UK Atomic Energy Authority Report, AEEW-R162.

Wallis, G. B. (1969) *One-dimensional two-phase flow*, McGraw Hill, New York.

Yasui, G. and Johnson, L. N. (1958) Characteristics of gas pockets in fluidized beds, *AIChE J.*, **4**, 445–452.