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HYDRODYNAMICS OF GAS-SOLID FLUIDIZATION

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Abstract—Work published on gas—solid fluidization since 1986 is reviewed, with emphasis on findings that appear to be new or to represent significant steps forward in advancing the understanding of fluidization phenomena, or which have potential practical implications. Hydrodynamic regimes ranging from bubbling to fast fluidization are addressed. Mixing phenomena and circulating fluidized beds are given special attention.

Key Words: fluidization, gas-solid systems, hydrodynamics, flow regimes, bubbling, circulating fluidized bed, downer, mixing

1. INTRODUCTION

Interest in fluidized beds has continued unabated in recent years, spurred on by new applications for fine particles and an ever-growing role for circulating fluidized beds and other high velocity systems. A large number of research papers, reports and other literature continue to appear presenting advances in fluidization, new applications and improvements in technology.

In order to keep this review to reasonable proportions, we confine ourselves to gas-solid systems. Liquid-solid fluidized beds lie outside our scope as they remain much less studied and of much less practical importance than gas-solid fluidized beds. Three-phase (gas-liquid-solid) fluidized beds are of increasing interest, but they have been the subject of an excellent book (Fan 1989). Although they are sometimes included in books or conferences on fluidized beds, we also exclude spouted beds in our purview since they tend to have a rather separate literature and are the subject of a separate review (Epstein & Grace 1995). Although they are often studied with fluidized beds and used in auxiliary equipment, we also treat neither solids flow in standpipes nor pneumatic conveying. Limited space has been available also for coverage of low-velocity systems. These subjects are covered in some detail in a recent book by Kwauk (1992).

In this review we consider work related to achieving an understanding of the hydrodynamics of gas-solid fluidized beds with special emphasis on high velocity systems and solids mixing. We do not treat heat or mass transfer or reactor modelling, even though these are closely dependent on hydrodynamics. There is extensive literature related to specific fluid bed applications. In the area of fluidized bed combustion alone, more than a hundred papers have appeared annually for at least the past decade. Except where such papers elucidate our understanding of fluidization behaviour, we also exclude this material from this review. In terms of the period covered, we have concentrated on literature that has appeared in print in 1986 or later. This seems a logical choice since two major texts (Davidson *et al.* 1985; Geldart 1986) covered work up to about that time.

Even with these exclusions, we have had to adopt a rather subjective approach to the coverage, given the volume of published work. We have featured subjects and findings that appear either to be new or to represent significant steps forward in advancing the understanding of fluidization phenomena, especially those which may have practical implications.

2. HYDRODYNAMIC REGIMES AND TRANSITIONS

Some advances have been made in recent years in characterizing different flow regimes and in predicting the transitions between them. It is frequently useful to apply the analogy with gas-liquid

two-phase flow in seeking to understand hydrodynamic regimes in gas-solid systems (Grace 1986). The hydrodynamic regimes of importance are shown schematically in figure 1.

2.1. Minimum fluidization

The lowest transition condition, the superficial velocity at minimum fluidization $U_{\rm mf}$, continues to be the subject of some investigation despite the considerable volume of previous work on the subject. Based on a balance of pressure drops required to support the weight minus buoyancy acting on the particles at the point of minimum fluidization and the well-known Ergun equation, most equations are of the form

$$\operatorname{Re}_{\mathrm{mf}} = \sqrt{C_1^2 + C_2 \operatorname{Ar} - C_1},$$
 [1]

where Re_{mf} and Ar are the Reynolds and Archimedes numbers given by

$$\operatorname{Re}_{\mathrm{mf}} = \rho_{\mathrm{G}} d_{\mathrm{p}} U_{\mathrm{mf}} / \mu_{\mathrm{G}}, \qquad [2]$$

$$Ar = \rho_{\rm G} \Delta \rho g d_{\rm P}^3 / \mu_{\rm G}^2, \qquad [3]$$

where $\Delta \rho = (\rho_p - \rho_G)$. Here ρ_p , ρ_G , d_p , μ_G and g refer to particle and gas density, particle diameter, gas viscosity and gravity respectively. New pairs of values of (C_1, C_2) have been proposed (Lucas *et al.* 1986; Adanez & Abanades 1991) which are particle-shape dependent and species dependent, adding to the large number of pairs already in the literature as summarized by the latter authors.

Much higher values of gas flow than predicted by purely hydrodynamic approaches are required when the temperature is raised to a point where agglomeration begins to occur (Yamazaki *et al.* 1986; Davies *et al.* 1989). Even when agglomeration is not a factor, great care is needed to measure U_{mf} at high temperature, in particular to ensure temperature uniformity (Flamant *et al.* 1991).

2.2. Minimum bubbling

Group A powders show an appreciable bubble-free velocity range between $U_{\rm mf}$ and the minimum bubbling velocity $U_{\rm mb}$. While there continue to be contrary views, supported by instability theory based purely on hydrodynamic considerations (e.g. Foscolo & Gibilaro 1987; Gibilaro *et al.* 1988;



Figure 1. Schematic diagram showing hydrodynamic regimes of fluidization (Grace 1986).

Batchelor 1988; Foscolo 1989), most fluidization researchers believe that interparticle forces play an important role for group A powders throughout the bubble-free range and in determining $U_{\rm mb}$. Hence, complex rheological behaviour and non-hydrodynamic factors appear to play significant roles, making predictions difficult (Homsy *et al.* 1992). There is also further evidence (Jacob & Weimer 1987; Foscolo *et al.* 1989) that both $U_{\rm mb}$ and $\varepsilon_{\rm mb}$, the voidage at the minimum bubbling condition, increase with increasing system pressure.

The condition for distinguishing group A powders from group B particles, i.e. the maximum particle diameter for $U_{\rm mb}$ to be appreciably greater than $U_{\rm mf}$, can be approximated (Grace 1986) by the empirical relation

$$d_{\rm p} \le 101 \{\mu_{\rm G}^2 / \rho_{\rm G} \Delta \rho g\}^{1/3} (\Delta \rho / \rho_{\rm G})^{-0.425}.$$
 [4]

2.3. Onset of turbulent fluidization

As interest in the higher velocity regimes of fluidization has blossomed, spurred by applications of turbulent and circulating fluidized beds, there has been increased investigation of the onset of these regimes.

Since early work by Yerushalmi & Cankurt (1979), the transition to turbulent fluidization has usually been characterized by the superficial velocity U_c at which the amplitude of pressure fluctuations reaches a maximum, or by the superficial velocity U_k at which the amplitude of pressure fluctuations levels off with increasing superficial gas velocity U. As summarized by Brereton & Grace (1992), there has been wide variation in the manner in which the experimental pressure fluctuations have been measured and analysed, some workers preferring absolute or dimensional values, while others employ differential values and/or normalize to give dimensionless values. U_k is not a well defined parameter since it depends, among other factors, on the solids return system employed (Bi & Grace 1995), and so its use is not recommended. The method used to determine U_c significantly affects the result (Bi & Grace 1995). Values in the extensive literature based on absolute pressure fluctuation data and bed expansion measurements are well represented by an equation due to Cai *et al.* (1989),

$$\operatorname{Re}_{c} = \rho_{\rm G} \overline{d}_{\rm p} U_{\rm c} / \mu_{\rm G} = 0.57 \operatorname{Ar}^{0.46},$$
[5]

while differential pressure fluctuation data are well represented by

$$Re_{c} = \rho_{G} d_{p} U_{c} / \mu_{G} = 1.24 Ar^{0.45}$$
[6]

(Bi & Grace 1995). These equations improve on several expressions available in the literature of similar form. Since the differential pressure fluctuation data are more indictive of local conditions, [6] is recommended. Transition data based on visual observations tend to give smaller values of the transition velocity and to be highly subjective.

The transition to turbulent fluidization corresponds to breakdown of bubbling or slugging due to rapid coalescence and splitting beyond a certain point (Cai *et al.* 1990; Bi *et al.* 1996). Accordingly, the transition may be affected by such factors as the presence of baffles (Andersson *et al.* 1989) and particle size distribution (Sun & Grace 1992), which affect the growth and breakup of bubbles. Transition can occur quite sharply for group A powders where the transition is from bubbling to turbulent, or much more gradually for group B or D particles where slugging occurs first and the transition involves intermittent periods of slug-like and turbulent character (Brereton & Grace 1992). Caution is needed when studying the transition to avoid an apparent transition caused by gradual emptying of the interval between two pressure taps over which differential pressure fluctuation measurements are being taken (Rhodes & Geldart 1986a).

2.4. Transition to fast fluidization

Bi et al. (1995) showed that the transition from turbulent to fast fluidization corresponds to a critical superficial velocity U_{se} which corresponds to the onset of significant particle entrainment



Figure 2. Dimensionless regime diagram corresponding to Bi et al. (1995) extended from that of Grace (1986).

from the riser. Except for columns of small size, U_{se} represents an equipment-independent property of the particulate material. Values of U_{se} are well correlated by

$$U_{\rm se} = 1.53 [g d_{\rm p} \Delta \rho / \rho_{\rm G}]^{0.5},$$
^[7]

which makes $U_{se} > v_T$ (the terminal velocity) for group A and B particles (especially the former), while U_{se} is essentially equal to v_T for group D particles. For $U > U_{se}$, the flow pattern of circulating fluidized beds depends on the solids circulation rate as well as U.

2.5. Transition to dilute pneumatic conveying

If the superficial gas velocity is raised through the fast fluidization regime at a fixed solids flux G_s or solids-to-gas loading ratio, one eventually reaches a gas velocity at which there is no accumulation of solids at the bottom. The point where solids accumulation commences on lowering the gas velocity corresponds to what has been called (Bi *et al.* 1993b) type A (or accumulative) choking. This superficial velocity is designated U_{cA} and is well predicted by an equation due to Bi & Fan (1991) which may be written explicitly as

$$U_{\rm cA} = 7.34 (gd_{\rm p})^{0.324} (G_{\rm s}/\rho_{\rm G})^{0.352} {\rm Ar}^{0.068}.$$
 [8]

This approach has been independently developed by Yang (1994) based on earlier work by Takeuchi et al. (1986).

2.6. Other choking modes

In addition to the accumulative choking mode given in the previous sub-section, there are two other phenomena commonly referred to as choking (Bi *et al.* 1993b). Type B (or blower/standpipe induced) choking occurs when the blower has insufficient capacity or the standpipe is of insufficient length to permit the system to operate with the required circulation rate of solids at the given gas velocity. Type C (or classical choking) corresponds to a transition to severe slugging in the transport line. Bi *et al.* (1993b) suggest approaches to predict these transitions which occur with decreasing gas velocity at a fixed value of circulation flux. The recognition that the term choking is commonly used to describe several quite different phenomena with different causes has helped to dispel the scatter and confusion which have long surrounded the literature on choking.

2.7. Regime diagram and powder types

The information covered in this section is summarized in figure 2, a regime diagram (Bi *et al.* 1995) in which a dimensionless superficial gas velocity

$$U^* = U[\rho_G^2/g\Delta\rho\mu_G]^{1/3}$$
[9]

is plotted versus $Ar^{1/3}$, a dimensionless particle diameter, the same coordinates as employed by Grace (1986). Note that the turbulent regime extends from the line for U_c , [6], to that given by [7] for U_{se} . The onset of dilute pneumatic conveying depends on the solids-to-gas volumetric flow rate ratio $m = G_s/\rho_0 U$.

The above regime diagram not only provides a way of showing which hydrodynamic regime is applicable, but it also shows the boundary between Geldart group B and D powders proposed by Grace (1986), i.e.

$$Ar = 1.45 \times 10^5,$$
 [10]

together with a typical CA boundary and an approximate AB boundary from [4], with $\rho_p/\rho_G \approx 1500$. Regime diagrams of this kind can be extended to continuous upward transport systems (Bi & Grace 1995a).

2.8. The fluidized bed as a chaotic system

There has been extensive work on chaos in a number of fields in recent years. It has been pointed out that the hydrodynamics of fluidized beds exhibit many features of chaotic dynamic systems in that they show behaviour which is both irregular and deterministic. Many properties of fluidized beds, such as local pressures, local voidage and local concentrations of chemical species undergoing reaction, show irregular, but non-random, fluctuations related to non-linear dynamics of the system. Experimental measurements (e.g. van der Stappen *et al.* 1993 and 1994; Bouillard & Miller 1994) confirm that the behaviour is chaotic and indicate that chaos analysis may provide useful tools to characterize the dynamics of fluidized beds operating in the various hydrodynamic regimes, with different distinguishing features in each regime.

Theoretical approaches are being pursued by several research groups. Daw & Halow (1992) have shown that deterministic chaos can arise from bubble interactions. Schouten & van den Bleek (1992) have taken a fundamental approach in which the irregular oscillating motion of five particles in a vertical one-dimensional array is treated, with inclusion of drag, gravity and interparticle collisions. Application of chaos theory to diagnostics and control of fluidization hydrodynamics has been reported by Daw & Halow (1993).

3. INTERPARTICLE FORCES AND INFLUENCE OF PARTICLE SIZE DISTRIBUTION

There are continuing attempts to extend fluidization to finer and finer particles, into the range of cohesive particles found in group C of the Geldart classification. At the same time, more attention is being given to effects of interparticle forces for both group C and group A powders.

A useful review of cohesive forces affecting fluidization has been published by Visser (1989). In some cases, these cohesive forces may be counteracted sufficiently by external means to allow fluidization to proceed, e.g. by an acoustic field (Chirone *et al.* 1993). It has also been found that some fine powders will, beyond a certain minimum superficial velocity, spontaneously form agglomerates which are large and stable enough to fluidize like a group A powder (e.g. see Brooks & Fitzgerald 1986; Li *et al.* 1990). More work is required to understand this behaviour and to devise practical techniques for extending fluidization to finer particles.

When bubbling fluidization does occur, Clift (1993) and Clift & Rafailidis (1993) have shown that interparticle stresses play a key role in bubble wakes while, for group A, B and D solids, interparticle stresses play a secondary role elsewhere and can often be neglected to a first approximation. In other words, the motion of bubbles, once formed, is insensitive to the rheological properties of the dense phase. Clift (1993) showed, on the other hand, that to explain the pre-bubbling differences between group A and B powders, one must consider interparticle forces, in particular the elasticity of the particulate phase which is critically dependent on particle-particle contacts. This is a very active area for future research.

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For many years industrial operators of catalytic fluidized bed reactors have recognized that it is important to maintain a significant proportion of 'fines', i.e. particles with diameters much smaller than the mean, within fluidized bed reactors in order to optimize reactor performance. The extent of the influence of fines and the underlying causes have become better understood in recent years.

Both Yates & Newton (1986) and Pell & Jordan (1988) demonstrated that addition of catalyst fines to fluidized bed reactors causes significant improvements in conversion, even when the added fines are catalytically inactive. In these two studies, the mean particle size, as well as the size distribution, were altered as fines were added. Sun & Grace (1990) performed experiments where the mean particle size [defined as $d_p = 1/\Sigma(x_i/d_{pi})$, where $x_i = \text{mass}$ fraction of size d_{pi}] was held constant, while three different size distributions were investigated. A wide size distribution always gave higher conversions (i.e. better gas-solid contacting) than a narrow size distribution, with a bimodal distribution showing intermediate results. In each of the cases above, the authors employed group A particles. Somewhat similar results were obtained by Kono & Soltani-Ahmadi (1990) by adding inert fines to group B particles undergoing a gas-solid reaction. Addition of finer particles usually led to higher conversions, as well as lower pressure fluctuations, suggesting smaller bubbles.

Geldart & Buczek (1989) found that bed expansion, entrainment and collapse test de-aeration times all increased significantly when the finest 3% by mass of particles (mean size $32 \mu m$) of FCC powder was removed and replaced by small quantities of ultrafine particles of various types. The extent of the increase was greater when the size of the added ultrafine material was diminished. A reduction in bubble size appeared to be responsible for the observed influence.

The improved performance of fluidized bed reactors with wide particle size distributions and appreciable quantities of fines is due to a number of factors (Grace & Sun 1991):

- (a) In the bubbling regime, voids tend to be smaller with wide particle size distributions (Hatate *et al.* 1988; Geldart & Buczek 1989; Soltani-Ahmadi 1989; Sun & Grace 1992), probably associated with lower effective dense phase viscosities (Khoe *et al.* 1991).
- (b) There are more particles dispersed inside the dilute phase when there are fines present (Sun & Grace 1990 and 1992), because fines spend more time inside voids when their terminal settling velocities are of similar magnitude to the relative throughflow velocity of gas inside the void (Grace & Sun 1990).
- (c) Wide size distributions trigger earlier transition to the turbulent fluidization regime where gas-solid contacting is better than in the bubbling regime.

These findings are primarily for group A powders. Work is required to see whether they are also applicable to group B and D solids.

4. BUBBLING BED HYDRODYNAMICS

Much has been written about bubbling phenomena in fluidized beds over the last three and a half decades. A good understanding of the bubble hydrodynamics is necessary to understand bubble-related phenomena such as solids mixing and segregation, reaction conversion, heat and mass transfer, erosion of heat transfer tubes and particle entrainment in beds operated in the bubbling regime. In this review, recent studies related to bubbling or slugging fluidized beds are briefly discussed.

Bubble size, velocity, shapes and flow patterns are of key interest in bubbling hydrodynamics. These properties have been extensively measured experimentally by various methods. The experimental methods and findings have been summarized in several review articles (Davidson *et al.* 1985; Geldart 1986; Cheremisinoff 1986). Here we review highlights of both experimental and theoretical studies at ambient conditions, and at elevated temperatures and pressures.

4.1. Experimental studies

Bubbles in a fluidized bed can be detected by measuring changes of local resistivity, capacitance, induction, radioactivity, pressure or optical attenuation. Numerous experimental studies have used immersed probes to determine bubble parameters. The results from such techniques are usually not sufficient to explain the intricate behaviour of bubbles or the scatter found when bubble velocity

is plotted against bubble size. Several new approaches for the determination of bubble shape have been considered recently to investigate these aspects in a greater depth.

Hatano *et al.* (1986) carried out spatio-temporal measurements of bubble properties in a three-dimensional free-bubbling fluidized bed using arrays of reflective optical fibre probes. Bubble size and shape can be resolved from the signals obtained by the assembly of probes. The probe array, arranged like two sets of combs with 13 probes in each line 10 mm apart, was inserted into a 0.15 m diameter column. Lim *et al.* (1990), Lim & Agarwal (1990 and 1992) and Mudde *et al.* (1994) utilized digital image analysis to determine distributions of bubble diameter, velocity, shape and angle of rise in two-dimensional beds.

Halow *et al.* (1990) and Halow & Nicoletti (1992) used a non-invasive rapid capacitance imaging technique to measure voidage, bubble and slug sizes, rise velocities, spacing and frequency. The capacitance was determined in 49 volumes at four adjacent vertical levels with sensing electrodes mounted flush with the walls of the column. Three-dimensional contour images of slugs were constructed from the system. The method has its limitations, such as blurring due to low resolution of the system (a pixel size of $10 \times 10 \times 25$ mm), non-linearity of void fraction versus permittivity due to electrical interaction between particles and anomalies in voidage measurement when two voids overlap at the same level.

Hatano *et al.* (1986) showed that the extent of bubble deformation was correlated with a 'coalescence index' defined by $(U - U_{mf})/U_b$, where U_b is the bubble velocity. The trailing bubble during coalescence elongated and had a higher rising velocity. When two bubbles rose side by side, they scarcely affected each other, although the bubble shape was slightly elongated. Halow *et al.* (1990) found that bubbles travelled faster when separated by less than 1 to 2.5 bubble diameters for fluidized beds of coarse nylon spheres and fine FCC particles. Both Hatano *et al.* (1986) and Lim & Agarwal (1992) distinguished different kinds of bubbles by plotting the rise velocity against bubble diameter. The distribution of velocities was narrower for single bubbles than for swarms due to bubble interactions.

Most studies used group B particles of Geldart's classification, except for Glicksman *et al.* (1987), Clark *et al.* (1991), Halow & Nicoletti (1992) and Luca *et al.* (1992), where group D particles having particles as large as 3.2 mm were used. Slugging was more predominant with the group D solids except for studies using large column cross-section. In large beds of coarse particles bubbles continue to grow without reaching any observable maximum bubble size, unlike group A powders (Geldart & Xie 1992). Clark *et al.* (1991) determined the slugging frequency from the autocorrelation function, Fourier transform and power density function and found that it decreased with bed height. The slug velocity increased with air throughput and decreased slightly with height. Luca *et al.* (1992) studied the influence of bed diameter on the transition from bubbling to slug flow conditions and correlated the main slug characteristics to physical properties and operating parameters using dimensionless groups.

In the presence of a tube bank (5 rows of 51 mm diameter tubes in a triangular pitch of 15 mm by 13.5 mm), Glicksman *et al.* (1987) found that individual bubble velocities were close to the value expected for isolated bubbles, while the bubble diameter did not increase appreciably with excess gas velocity, remaining 1 to 1.5 times the tube pitch.

Since most industrial bubbling fluidized bed reactors operate at elevated temperatures and pressures, knowledge of how these parameters affect bubble properties is important. Knowlton (1992) provided a good overview of the pressure and temperature. The change in gas density and viscosity caused by temperature and pressure affect such properties as minimum fluidization velocity, bed voidage and expansion, bubble size and frequency, entrainment and regime transitions. Recent evidence suggests that bubble size decreases slightly with temperature for group A powders (Hatate *et al.* 1988a, 1988b and 1990) but to a lesser extent for coarser particles (Sishtla *et al.* 1986; Weluda *et al.* 1987). Measurements from Botterill & Hawkes (1992), however, showed a small increase of bubble size with temperature for 1.23 mm sand particles.

There is also further experimental evidence that increasing pressure leads to smaller bubbles resulting in smoother fluidization. Chan *et al.* (1987), Piepers & Rietema (1989), Foscolo *et al.* (1989), Cârsky *et al.* (1990) and Hatano & Kido (1992) have covered a range of pressures up to 5 MPa. Chan *et al.* (1987) found that $U_{\rm mf}$ decreased with pressure while $U_{\rm mb}$ increased with increasing pressure. They also observed a 78% reduction in bubble size for 0.1 mm particles

compared with a 32% reduction for 0.4 mm particles when the pressure increased from atmospheric to 3.1 MPa. Bubble frequency also increased with pressure, especially for the finer powder. It is believed that a group A powder experiences an increased dense phase expansion at high pressure causing a lower effective dense phase viscosity. Taylor instability increases with decreasing dense phase viscosity, thereby promoting more splitting from the bubble roof, leading to smaller bubbles. Piepers & Rietema (1989), on the other hand, studied the fluidization behaviour of fresh cracking catalyst and polypropylene powder up to 1.5 MPa with 6 different gases. The maximum homogeneous bed voidage ε_{mb} and the minimum bubbling velocity U_{mb} increased with pressure, with the increase strongly affected by the gas used. The bubble size (inferred from bed collapse tests) was shown to decrease with increasing pressure. However, Hatano & Kido (1992) found that there was no clear effect of pressure on bubble growth for FCC particles with pressure up to 5 MPa. More work is needed to determine the effect of temperature and pressure on bubble phenomena.

4.2. Measurement with probes

Submersible probes are often used to study local bubble properties in gas-fluidized beds. Since the axes of the intercepted bubble need not be aligned with the axis of the probe, simple probes measure only the pierced length of bubble and the determination of bubble diameters requires care. The pierced length distribution P(y) is not explicitly related to the actual bubble size distribution P(R), where R is the characteristic bubble dimension, e.g. bubble horizontal radius. The two distributions are related by:

$$P(y) = \int_0^{R_{\text{max}}} P(y|R) P(R) dR,$$
[11]

where P(y|R) is the conditional probability of measuring a pierced length y from a bubble characterized by R integrated over a possible range of bubble sizes (0 to R_{max}) having distribution defined as P(R). In general, the conditional probability P(y|R) is related to the spatial uniformity of the bubbles within the region of detection of the probe and the geometrical shape of the bubble. The relationship between measured chord lengths and true bubble sizes of uniform size distribution for several typical bubble shapes (spherical, ellipsoidal, spherical and ellipsoidal caps, etc.) has been illustrated using a forward transformation method (Clark & Turton 1988). A backward transform method, more useful in practice, has been formulated to convert distributions of pierced lengths to bubbles sizes by discretizing the integral in [11] (Clark & Turton 1988; Turton & Clark 1989 and 1992). However, this method may become unstable, leading to severe inaccuracy when there are too few data points or the discrete size intervals of the distribution are too many. Very irregular (multimodal) bubble size distributions may occur in practice and these can be difficult to treat using the above backward transform.

Lim & Agarwal (1990) conducted studies on a 2-dimensional bed using image analysis to obtain simultaneously the pierced length and several size measures of bubbles intercepted by an imaginary probe for several operating conditions. The measured pierced lengths, together with assumed bubble shapes, were used to obtain theoretical prediction of the bubble size measures, using a geometrical probability approach involving the backward transform method proposed by Clark & Turton (1988). To convert the experimental pierced length to bubble size, Lim & Agarwal (1990) fitted the pierced length distribution with a continuous Gamma distribution function. A range of bubble size measures (bubble horizontal, vertical and area-equivalent diameters, circumference) predicted by the above method for four typical bubble shapes, (spherical, ellipsoidal, spherical cap and hemispherical) were compared with experimental data. Spherical and ellipsoidal bubble shapes gave good agreement. They also suggested two refinements to strengthen the method. It was confirmed experimentally that the assumption of homogeneity in bubbling behaviour used in the theory may not be strictly correct. The theory also assumes a uniform shape for the entire bubble population. However, due to complex interactions between neighbouring bubbles in a swarm, bubbles have various shapes. Undoubtedly inclusion of one more size characteristic (e.g. aspect ratio) would demand an additional stage of integration, ultimately requiring a more sophisticated algorithm.

Extensive computational effort is required to convert measured pierced lengths to true bubble sizes. The characteristic bubble shape must be known *a priori*, which can be difficult. When the actual size distribution is not required, the mean bubble size can be assumed to be proportional

to the mean value of pierced length measured by the probe. Lim & Agarwal (1990) have shown experimentally that the average bubble size d_b can be correlated with measured mean pierced length $\xi(y)$ in the form $d_b \approx 1.35 \,\xi(y)$. This relation is of the same form as that commonly used by other workers (Gunn & Al-Doori 1985; Weimer *et al.* 1985; Chan *et al.* 1987) where d_b varies from 1.2 to 1.5 $\xi(y)$.

Bubble velocity is calculated from the time delays between two channels, either by a simple cut-off method or via a more elaborate cross-correlation approach. Very often the pulses are not rectangular or regular and two pulses do not give the same values of velocity and pierced length. Geldart & Xie (1992) attributed these problems to: (a) the effect of signal fluctuations; (b) the elongation and acceleration of bubbles as they pass a probe; (c) bubbles rising obliquely, passing the lower but not the upper probe, or *vice versa*; (d) the splitting of bubbles after they have passed the lower probe.

Lim & Agarwal (1992) showed that the angle of bubble rise must be taken into account in interpreting signals, especially for dual-tip probes. Procedures should be incorporated in probe designs to detect and reject non-vertically rising bubbles. Analysis of the data by Lim & Agarwal (1992) established that rejection of non-vertically rising bubbles (e.g. Sung & Burgess 1987) does not significantly bias the measurement of bubble characteristic. Multi-element probes should therefore be used rather than dual-element probes whenever possible.

4.3. Gas flow inside bubbles

Because of the pressure gradient across voids in fluidized beds, gas flows upwards through the voids from bottom to top. This upward flow is important in stabilizing the upper surface of bubbles, in contributing to dilute-phase-to-dense-phase mass transfer of gas and in accounting for the division of gas between the dense and dilute phases. It is therefore of considerable importance to know the magnitude of the flow relative to the bubble, commonly called the 'throughflow'.

Throughflow is very difficult to measure in fully three-dimensional fluidized beds, although some measurements from Hilligardt & Werther (1986), employing a combined capacitance-pressure probe, indicate a throughflow velocity U_{tf} of order 2.7 U_{mf} for three-dimensional columns. Several studies have been reported in recent years in which throughflow gas velocities were measured using (non-intrusive) laser Doppler anemometry (LDA) in two-dimensional columns. Previous measurements (e.g. Garcia *et al.* 1973) had only established that the throughflow velocity (i.e. the mean gas velocity inside the void relative to the void) is of the same order as the minimum fluidization velocity, in accordance with theory (e.g. Murray 1965).

Gautam *et al.* (1991) performed experiments with glass beads of mean size 350 and 500 μ m, with the beds fluidized at $U/U_{mf} = 1.05$ and 1.13, respectively, these values providing bubbles of stable size. They then injected periodic pulses of air to form large bubbles. Strobe flashes and videotaping allowed the bubble velocities to be measured and then subtracted from the local gas velocities determined by forward scatter LDA. Bubbles were found to elongate as they rose and this probably explained why the measured throughflow velocities were found to increase with increasing height. In the case of the 350 μ m particles, U_{tf} varied from 1.4 U_{mf} to 1.7 U_{mf} between heights of 170 and 250 mm above the distributor, whereas the variation was from 1.38 U_{mf} to 1.46 U_{mf} for the 500 μ m particles over the same height interval. In both cases, the throughflow velocity appeared to be independent of horizontal and vertical position within the bubble at a given height. Good predictions of these U_{tf} values have been obtained by an analysis (Gera & Gautam 1994) which makes allowance for both bubble deformation and voidage variations outside the bubble boundary similar to those measured experimentally by Halow & Nicoletti (1992) and Yates *et al.* (1994).

In another study, Hailu *et al.* (1993) used a similar technique, except that back scatter was used instead of forward scatter for the LDA measurements. In this case the fluidized particles were sand of mean diameter $450 \,\mu$ m, while the background superficial velocity was maintained at $U/U_{\rm mf} = 1.00$ and the bubbles were recorded by cine photography. Fine alumina particles were injected into the bed together with the air pulses forming the bubbles in order to trace the gas motion inside the bubbles. $U_{\rm tf}$ was found to increase with bubble size, possibly due to bubble elongation caused by the side wall. In a similar way to Gautam *et al.* (1991), they found that $U_{\rm tf}$ increased with height, although in the case of Hailu *et al.* (1993), the increase seemed to be only as the bubbles approached the bed surface where an increase in throughflow is expected theoretically (Yule & Glicksman 1988). The ratio U_{tf}/U_{mf} ranged from about 0.8 in the middle of the bed to about 1.8 near the bed surface. The latter value is close to a value of the ratio of 1.84 reported for two-dimensional beds by Hilligardt & Werther (1986).

The differences between these ratios measured in the different studies could reflect the different $U/U_{\rm mf}$ values used, differences in particle shape reflected in different voidages near the bubbles, or different bubble distortion due to wall effects. Further experimental work is needed to clarify these differences and to allow more complete comparisons with theoretical throughflow velocities.

4.4. Hydrodynamic modelling

In recent years, there has been a strong focus on semi-empirical and theoretical modelling of bubbling fluidized beds. Choi *et al.* (1988) developed a theoretical model for mean bubble size and frequency based on collision theory, assuming a random spatial bubble distribution in freely bubbling fluidized beds. A correlation was derived which gives good agreement for a larger number of data from the literature, as well as their own measurements, for $d_p = 0.041$ to 1.760 mm, D = 0.10 to 1.22 m, U = 0.009 to 2.14 m/s, T = 20 to 1000°C and P = 1 to 71 bar. Under specific conditions, the above correlation was shown to reduce to earlier correlations. Agarwal (1985 and 1987) used a population balance to calculate the bubble size distributions. This model assumes bubble growth by coalescence, with bubble breakage neglected. It is therefore only applicable to Geldart group B and D particles.

Horio & Nonaka (1987) derived a new correlation to predict bubble diameters as a function of height in beds of non-cohesive powders (groups A, B and D) in contrast to earlier correlations which apply only to group B and D particles. Bubbles were allowed to reach a maximum size at some height above the distributor for group A powders due to a balance between coalescence and splitting processes. The coalescence frequency was estimated from data for type B and D powders where negligible splitting occurs. The splitting frequency was assumed to depend on particle diameter but not bubble diameter, with splitting frequency estimated from other experimental results. The correlation covers a range of gas velocity (0.05 to 0.5 m/s) and bed diameter (0.079 to 1 m) with $\pm 20\%$ uncertainty. Good agreement was obtained with previous experimental data. More fundamental work on bubble splitting is needed to achieve more reliable predictions.

Gidaspow et al. (1986) computed formation, growth and bursting of a single bubble in a two-dimensional fluidized bed produced by a gas jet from a complex hydrodynamic model. The results compared well with experiment. Qualitative representation of the mixing particles of two sizes was also generated by supercomputer calculation. Syamlal & O'Brien (1989) extended the work of Gidaspow et al. (1986) and made quantitative comparison with experimental data. It was demonstrated that bubble formation, coalescence, motion and gas and solids mixing caused by bubbles can be satisfactorily described by a two-phase hydrodynamic model. Rafailidis et al. (1991) investigated the effect of distributor characteristics on bubble motion, simulated using the mechanistic model of Clift & Grace (1970 and 1971). Standpipe distributors were predicted to lead to localized areas of intense bubbling inside the bed which might lead to localized wear of tube bundles. Under some circumstances, tube wastage might be alleviated by increasing the number of bubble formation sites on the distributor. Though the above approaches provide sophisticated techniques for diagnosing distributor design and for scale-up without recourse to empirical methods, the computational power required is commonly not warranted for practical fluidized bed design. Further work on these fundamental models is, however, strongly recommended. Theoretical representation of bubbling phenomena using chaos theory should also be explored.

5. SOLIDS MIXING, SEGREGATION, PARTICLE MOTION AND EROSION

A good understanding of solids mixing behaviour is important in the design of physical and chemical processes in bubbling fluidized beds. Gas mixing, though related to solids mixing (Bellgardt *et al.* 1987), is not considered in this section.

Solids mixing has been reviewed by Potter (1971), van Deemter (1985), Fan *et al.* (1990) and Kunii & Levenspiel (1991). It is well recognized that solids mixing is directly related to bubble flow phenomena. The combined effects of gross circulation caused by drift and wake transport and small scale local-mixing in bubble wakes leads to favourable axial mixing. The extent of lateral solids

mixing is much less favourable, particularly in shallow fluid beds (height to diameter ratio H/D < 0.25) where the influence of the axial wake transport is weakest.

Particle segregation due to differences in particle size, density or shape, though commonly considered separately, can be treated as a subset of mixing processes with inclusion of preferential particle settling effects. In recent years, greater attention has been directed to the study of individual particle motion in fluid beds.

5.1. Experimental studies

Experimental techniques used to study solids mixing, segregation or individual particle movement commonly require the use of tracer particles. Some typical tracer characteristics used recently include:

- (a) Magnetism (Avidan & Yerushalmi 1985),
- (b) Chemical differences (Berruti et al. 1986),
- (c) Sublimation (Bellgardt & Werther 1986),
- (d) Heat (Valenzuela & Glicksman 1984; Fan et al. 1986),
- (e) Colour (Kozanoglu & Levy 1991; Lim et al. 1993),
- (f) Radioactivity (Lin et al. 1985; Lyczkowki et al. 1993),
- (g) Positrons (Simons et al. 1993),
- (h) Fluorescence (Morooka et al. 1989).

Some novel techniques listed above have been detailed in the recent review by Yates & Simons (1994). In (a) to (e) above, the experimental results were presented as spatial concentration profiles, as transient responses or in terms of a mixing index, while methods (f) to (h) enabled measurement of the motion of individual particles and determination of time-average solids velocities.

Conventional methods of measuring solids mixing such as bed slumping and sectioning tend to be extremely time-consuming and tedious. Although tracer response can be continuously monitored by methods (d) and (f), investigations have usually been concerned with only the dense phase in isolation. Data delineation from bubble hydrodynamics has been difficult, in some instances leading to distorted data comparison. Lim *et al.* (1993) overcame some of the limitations by a digital image analysis technique which determined tracer concentrations continuously in a two-dimensional fluid bed in the presence of both the dense and bubble phases.

5.2. Solids mixing

A number of mathematical models have been proposed to predict mixing behaviour in bubbling fluidized beds. The need for a more realistic model based on relevant hydrodynamic parameters of bubbling beds was stressed by van Deemter (1985).

A one-dimensional diffusion model has been demonstrated to be inadequate in describing axial mixing behaviour due to observed cycling in the concentration responses (de Groot 1967; Lim *et al.* 1993), especially for large particle systems. Attempts have also been made to model axial solids mixing as a stochastic process (Fox & Fan 1987). However, lack of suitable experimental data for model verification has restricted the applicability of this approach. However, Fan *et al.* (1986) showed that lateral mixing can best be based on a stochastic diffusive model in which particle motion is characterized by both diffusive and convective components.

The counter-current back-mixing (CCBM) model originally proposed by van Deemter (1961) and refined and generalized by Gwyn *et al.* (1970) has gained greater acceptance due to its good representation of the transport process in a bubbling bed. The model depicts the bed as a multiple phase system, with an upward flow of gas and wake phases and a downward flow in the dense phase. Exchange occurs between these phases. Mass balances over the individual phases were represented by a system of hyperbolic partial differential equations. Some of the main features of the CCBM model have been adopted by various researchers. For example, Sitnai (1981) used this approach to model solids mixing in a fluidized bed containing horizontal tubes by including a fast moving, narrow downflow of solids at the wall. Numerical solutions through numerical inversion by Laplace transforms and a 'cinematic' approach have been provided by Lakshmanan & Potter

(1990). This method was shown to be more computationally efficient and robust than the former for impulse and pulse inputs. Verification of the CCBM model has been impaired until recently by a lack of experimental data on mixing and independent measurement of relevant bubble hydrodynamic parameters.

Kozanoglu & Levy (1991) developed a CCBM model which further divides the wake phase into four different compartments. Solids exchange between different phases is allowed except for the innermost wake phase which was taken to be stagnant. This multiple-layer wake region derived from an experimental/theoretical study by Kocatulum *et al.* (1992), where a strong particle velocity gradient in the wake was shown to exist. The nearly stagnant zone in the wake was believed to cause particles to be transported through large vertical distance. The solids exchange parameters between phases were adapted from the same study. The model provided a reasonable prediction of the axial concentration profile in comparison with experimental data.

Transient concentration responses measured by Lim *et al.* (1993) have been interpreted using a three-phase CCBM model similar to that of Gywn *et al.* (1970). Methods of predicting the wake exchange coefficient k_w , available in the literature (Yoshida & Kunii 1968; Chiba & Kobayashi 1977), were found to be inadequate. The wake exchange, estimated from fitting to experimental data, was found to be weakly influenced by the minimum fluidization velocity, contrary to the proposed correlations. The solids exchange coefficient was found to be related to bubble size with $k_w = A_w/d_b$, where A_w was of the order of 0.03 to 0.15 m/s for U_{mf} from 0.068 to 0.35 m/s.

This finding was further supported by Basesme & Levy (1992), where the wake exchange coefficient was measured in a two-dimensional bed using tracer displacement techniques and the data analysis method described by Chiba and Kobayashi (1977). The model of Chiba and Kobayashi (1977) agreed well with the experimental data over only a very narrow range of U_{mf} and d_b . The model developed by Kocatulum *et al.* (1992) overpredicted the exchange coefficient by a factor of two to close to an order of magnitude, depending on the range of U_{mf} and bubble size. The discrepancy was attributed to the theoretical analysis of Kocatulum *et al.* (1992) and the well-mixed assumptions in obtaining exchange coefficients from tracer measurements. The assumption of spherical or circular bubble wakes may also cause errors. The inability of the steady state model to account for the instabilities that lead to periodic wake shedding should also be taken into consideration. Hoffman *et al.* (1993) also pointed out that the proportionality of the solids exchange parameter to the minimum fluidization velocity did not hold. A better fit of the experimental data was realized when the dependency on U_{mf} was dropped.

The CCBM model is probably the best existing model to represent mixing in bubbling beds despite the fact that other possible mixing mechanisms, such as solids splashing at the bed surface and turbulent mixing near the distributor, are not accounted for. Although the solids convective component used in the model is well established, the key limitation which prevents application of this model with greater confidence is the absence of reliable solids exchange coefficients. Available experimental data are plotted against bubble size in figure 3. The corresponding minimum fluidization velocities varied from 0.004 to 0.35 m/s, while almost all of the exchange coefficient values are within the range estimated by Lim *et al.* (1993). Quantitative understanding of solids mixing in the wake region remains poor; more work is clearly needed to develop a more reliable prediction for the exchange coefficient.

5.3. Segregation

Segregation can arise in a bubbling fluidized bed containing a range of particle sizes and/or densities. Small and/or light particles ('flotsam') tend to migrate upwards while others ('jetsam') travel in the opposite direction. The degree of separation of the different components depends on the relative sizes as well as the relative densities of the fluidized particles. As with mixing, segregation is linked to the motion of bubbles in the fluidized bed. A dynamic equilibrium is set up between mixing and segregation under given fluidization conditions, leading to a characteristic axial concentration profile of flotsam/jetsam particles along the bed height. Studies related to segregation were reviewed by Nienow & Chiba (1985).

Fluidized beds displaying segregation can be broadly classified into two categories, namely jetsam-rich and flotsam-rich. Good overall solids mixing is desired in the fluidized bed combustion or gasification of solid fuels (such as coal, sawdust and straw). Solids mixing/segregation

characteristics in such applications have been studied by Ho *et al.* (1987), Hemati *et al.* (1990), Bilbao *et al.* (1988 and 1991) and Aznar *et al.* (1992). In coal cleaning processes, segregation is strongly promoted when magnetite particles are used as host particles (Kozanoglu *et al.* 1993).

The extent of segregation is commonly characterized either in terms of a 'macroscopic' mixing index that varies between 0 (complete segregation) and 1 (complete mixing), or by the axial concentration profile of the respective species. The mixing index is related to the superficial gas velocity and 'takeover velocity'. Empirical correlations for the takeover velocity include the influence of particle size, shape and densities, the relative proportion of jetsam and flotsam, and the bed aspect ratio (Rice & Brainovich 1986). Nienow et al. (1987) extended the segregation study from binary systems to quaternary systems in beds of different size particles. They found that the segregation patterns for a binary system of particles having equal density and unequal size are similar to those for binary systems with a density difference if the mixing index is based on the Sauter mean diameter rather than the mass fraction. Peeler & Huang (1989) provided an improved correlation for the takeover velocity for equi-density systems at high particle diameter ratios. Nienow et al. (1987) investigated the effect of distributor design on segregation and concluded that perforated plate and standpipe distributors provide better mixing than a porous plate at the same superficial gas velocity. The mixing index approach has also been discussed by Daw & Frazier (1988) who proposed a more general functional relationship between the mixing index and superficial gas velocity by incorporating the segregation index concept based on axial composition variance and a second velocity term to account for increasing segregation at high gas velocities. Delebarre et al. (1994) investigated the implication of various indices on the segregation state and introduced an index based on pressure drop along the bed height to give an on-line indication of the presence of segregation during fluidized bed operation.

The Gibilaro-Rowe model for segregation (see Gibilaro-Rowe 1974) has been adopted by many recent investigators to interpret steady (Garcia-Ochoa *et al.* 1989; Bilbao *et al.* 1988; Hoffman *et al.* 1993) and transient (Valkenburg *et al.* 1986; Bilbao *et al.* 1991) concentration profiles in a segregating fluid bed. The generalized model depicts the bed in terms of a bulk (dense) phase and a dispersed wake phase in which the solids flows are controlled by four mechanisms: convective, segregating, dispersive and particle exchange. Schouten *et al.* (1988) considered a similar approach to describe segregation in a slugging fluidized bed combustor of large particles. The solids flow



Figure 3. Relationship between solids exchange coefficient k_w and bubble diameter d_B obtained from experimental results in the literature.

mechanisms were based on downward solids flow in gas slugs, upward solids flow in particle slugs and exchange between upward and downward moving solids flows. Though the Gibilaro-Rowe model assumes that the segregation term is associated with the dense phase, Yoshida *et al.* (1980) consider it more likely to be in the wake phase. Retention of an axial dispersive term was required by Daw & Frazier (1988), while Nienow & Chiba (1985) considered it unnecessary.

Limitations of the Gibilaro-Rowe model discussed by Nienow & Chiba (1985), such as its inability to cope with a defluidized zone and poor prediction for systems with large differences in $U_{\rm mf}$, remain unexplained. Additional mechanisms of 'overlayering' in cases of high jetsam concentration systems should be considered.

Approaches similar to that used for solids mixing based on the CCBM concept have been adopted to model solids motion in a segregating fluidized bed by inclusion of an additional term describing the segregation propensity Y_s . The parameter Y_s is usually correlated to particle size and density ratios. The number of correlations available in the literature is presently limited (Hoffman & Romp 1991; Kozanoglu & Levy 1992). All correlations show a weaker influence of particle size on segregation than of density difference. The correlation of Hoffman & Romp (1991), a modified form of an expression due to Tanimoto *et al.* (1980), can be adequately used to estimate the segregation parameter. Theoretical and experimental results on the transient mixing/segregation behaviour of binary solids in a bubbling fluidized bed have been presented (Kozanoglu & Levy 1992; Lim 1992; Kozanoglu *et al.* 1993). Comparison of model predictions with the data of Lim (1992) indicates that, though these models did reflect some features of the experimental results, the influence of the superficial velocity on the temporal variation of concentration at any specific height within the bed is not well predicted. A trend completely opposite to that predicted was observed experimentally. No adequate reason was found for the poor agreement.

Beeckmans & Stahl (1987) considered a simple two-compartment model with interchange of particles to analyse kinetic data on the rate of segregation and desegregation in a strongly segregating system of iron and glass particles. The assumptions of a lower stratum of pure jetsam and a homogeneous upper stratum may not, however, be applicable for weakly or moderately segregating beds.

Other aspects relating to segregation have also been investigated, such as the use of a tapered fluidized bed to avoid segregation by maintaining a higher gas velocity near the inlet (Toyohara & Kawamura 1992). Bemrose *et al.* (1989) exploited segregation behaviour for ash management in a combustion system by exploring an angled multiple-compartment distributor with variable air distribution. An alternative method for enhancing segregation by means of stirring has been investigated by Zhang & Beeckmans (1990). The separation of binary mixture of particles in a continuously operated fluidized bed (Chiba *et al.* 1986; Vesely *et al.* 1991) and lateral mixing/segregation in beds of low aspect ratio (Milne *et al.* 1989; Milne & Nienow 1992) have also been of interest in recent years.

Understanding of segregation in bubbling fluidized beds is far from complete. Although reasonably reliable correlations are available for estimating the minimum fluidization velocity of a mixture (Chyang *et al.* 1989) needed to estimate bubble parameters in solids mixtures, in-depth understanding of bubble behaviour and solids flow phenomena in multi-species systems remains important. Initial results on the bubble behaviour in a binary system have been reported by Kage *et al.* (1991). Since most of the studies have been conducted in binary systems, more experimental work is clearly needed in view of the increasing number of applications of fluidized beds operating with a wide distribution of particle properties.

5.4. Particle motion

A good knowledge of the particle movement in fluidized beds assists in the understanding of solids mixing, erosion of immersed surfaces and the determination of the optimum location for feeding and withdrawing solids in fluidized beds. Particle motion may be measured using various tracer techniques as discussed above. Non-tracer techniques, such as strain gauges or force probes, have also been developed to measure circulatory motion of solids in a 0.96 m diameter fluidized bed (Yoshioka *et al.* 1987) and a 3 m diameter jetting fluidized bed (Ettehadieh *et al.* 1988). Unlike more sophisticated tracer tracking methods, the solids velocities were inferred from the force pulses. Yoshioka *et al.* (1987) found that the particles moved predominantly upward in the central core

(with dimensionless radial position r/R < 0.69) and downward in the outer annulus. Ettehadieh *et al.* (1988) identified circulation zones at the base of a tapered fluidized bed to consist of a transition zone sandwiched between upward core and downward wall zones.

Experimental work has also been extended (Rios *et al.* 1986; Lim & Agarwal 1994) to the study of circulatory motion of large light ('active') particles in a fluidized bed of smaller heavy particles, simulating coal particles in fluidized bed combustors. Particle properties and gas velocity are important in determining the circulation pattern, residence time and penetration depth of such active particles which were found to be increasingly associated with the bubble phase as the excess gas velocity was increased.

The importance of the solids movement has led to several modelling attempts. Soo (1986 and 1989) calculated the average circulatory motions of the solids in two- and three-dimensional fluidized beds by solving average dynamic equations of multiphase flow in the absence of bubbles. Despite criticisms by Gogolek (1991) on the formulation and the uncertainty in the choice of model parameters, the primary solution showed ascent at the centre and descent near the wall as the stable mode of bed particle motion in the upper region of the bed. The flow pattern of bubbles was suggested to be a consequence of particle circulatory motion rather than the cause of the solids circulation.

Lyczkowski *et al.* (1993) employed a computer-aided particle tracking technique to measure the velocity of a single radioactive tracer in a two-dimensional fluidized bed $(0.4 \times 0.038 \text{ m})$ containing two rows of round cylinders (51 mm diameter) in a triangular pitch arrangement with 152 mm horizontal and 76 mm vertical spacings. The experimental results compared reasonably well, in terms of overall solids circulation pattern and pressure fluctuations, with theoretical predictions from a hydrodynamic model based on first principles, an extension to that discussed in section 4.4 above. However, the solids velocities in the vicinity of the tube were overpredicted and possible reasons for the deviations were suggested.

Tsuji *et al.* (1993) developed a different modelling approach, a 'distinct element method', which expressed the contact forces between particles using springs, dashpots and friction sliders, taking into account interaction with fluid motion to simulate numerically the motion of discrete particles in a single nozzle two-dimensional fluidized bed. This approach was claimed to require fewer assumptions than a two-fluid model where the parameters affecting interparticle contact were determined from material properties such as Young's modulus, the Poisson ratio and the coefficient of restitution. In spite of the oversimplified assumption of negligible contact force between wall and particles, the predictions compared well with quantitative experimental data on pressure fluctuations and qualitative solids flow patterns.

The numerical modelling approach is elegant and should permit the effects of particle size distribution and density distribution to be analyzed. However, this approach is of limited application for fine (e.g. groups A and C) powders where the computational task will become very time consuming. Additional experimental work is clearly needed to validate the models.

5.5. Erosion

Erosion in the context of this review is a second-order effect primarily caused by gas and solids hydrodynamics. Numerous studies have been initiated because of the importance of erosion in combustion. A brief overview of recent erosion results (mainly on heat exchanger tubes) is presented here with emphasis on the role of fluidized bed hydrodynamics.

The scope of the study on erosion extends from measurement of tube wastage rate to detailed modelling of the mechanisms of erosion. Erosion rate measurements have been conducted both at room temperature and at high temperature by measuring the amount of tube material removed after immersion in a fluidized bed for a specified time. From a series of experimental findings (Holmes *et al.* 1989; Rathbone *et al.* 1989; Tsutsumi *et al.* 1989; Dennis 1989; Zhu 1988; Zhu *et al.* 1989, 1990 and 1991), key factors influencing the rate of erosion can be summarized into two general categories.

- (a) Bed hydrodynamics and properties:
 - (i) particle impact velocity (related to bubble velocity, size and hence operating gas velocity),

- (ii) particle impact angle,
- (iii) particle properties (size, density, sphericity, hardness),
- (iv) bed height and distributor type.

(b) Tube configurations and properties:

- (i) axial and lateral locations of tubes in the bed,
- (ii) tube bank configuration (pitch, spacing and orientation),
- (iii) tube and material properties (diameter, hardness, Young's modulus).

Although all these factors affect the overall erosion rate to various degrees, it is the particle movement that causes direct damage to the tube. The bottom half of heat transfer tubes generally suffers the most, with a maximum erosion rate about halfway between the bottom and sides, i.e. at about the 4:30 and 7:30 positions (Dennis 1989; Tsutsumi *et al.* 1989; Zhu *et al.* 1990). Rathbone *et al.* (1989) concluded, from the results of simultaneous measurement of the transient normal and tangential components of particle velocity and stress against a surface, that surface damage on the tube is caused by 'impact erosion' as well as 'abrasion wear' resulting from oblique impact of particles in the bubble wakes. The effects of various tube bank configurations on overall and local erosion depend on bed operating conditions. Tubes inside tube bundles are generally subject to less erosion (Zhu *et al.* 1990). The erosion rate tends to be higher for large group D particles (Rathbone *et al.* 1989; Zhu *et al.* 1990) than for smaller particles, and for higher gas velocities than for lower ones (Zhu *et al.* 1990). Hydrodynamic considerations (Levy & Bayat 1989; Zhu *et al.* 1989; Bayat *et al.* 1990; Levy *et al.* 1992) suggest that the most energetic impacts occur during bubble coalescence.

The extent of in-bed tube erosion rate may be characterized either by a semi-empirical correlation (Zhu *et al.* 1991) which includes the key variables and conditions, or by resorting to more sophisticated numerical methods which calculate the particle velocity distribution around the tube (Bouillard *et al.* 1989; Padhye *et al.* 1989; Bouillard & Lyczkowski 1991; Ding *et al.* 1992). The erosion rate can be related to the particle velocity by various erosion models based on energy dissipation. Although the kinetic collision energy of the particles has been studied by Kono *et al.* (1987 and 1989) using tracer particles of known mechanical strength, the knowledge of the energy distribution among different energy transfer mechanisms is at present limited. Yates (1987) proposed that erosion of a metal surface is directly proportional to the kinetic energy of particles in the bubble wake. However, Zhu *et al.* (1991) argued that only one or two layers of the particles in the wake can effectively impact on the bottom of the tube causing erosion. An approach which assumes energy dissipation of a mono-layer of particles at the tube surface has been proposed by Bouillard & Lyczkowski (1991).

Future work is clearly required to improve the numerical models with more detailed study on the erosion mechanism. Additional studies on the interaction between gas bubbles and immersed tubes (Yates & Ruiz-Martinez 1987) may also be useful. Novel tube geometrics which reduce erosion problems should also be further explored.

6. PARTICLE ENTRAINMENT AND ELUTRIATION

At high superficial gas velocities, fine particles can be effectively elutriated or removed from a fluidized bed. This is common in fluidized bed reactors with wide particle size distributions, or where fines are generated from particle attrition, combustion or reaction. As discussed in section 3, loss of fines from fluidized beds can adversely affect the reactor performance. It can also lead to loss of catalyst and air pollution. Therefore, the ability to determine entrainment rates and to capture and return entrained particles to the bed are critical in the design of fluid bed reactors.

Solids holdup in the freeboard typically decreases approximately exponentially with increasing height. Practical design considerations suggest that the gas exit be located above the transport disengagement height TDH [or TDH(F), Geldart 1986], defined as the height at which the solids flux or holdup becomes constant. Predictive models for the TDH are generally semi-empirical

(Geldart 1986; Kunii & Levenspiel 1991). Baron et al. (1988a) determined the TDH by considering the trajectory of clusters ejected into the freeboard.

Entrainment rates above the TDH, expressed as limiting entrainment rate constants κ_i^* have been correlated by various dimensionless parameters involving particle and fluid properties and operating conditions (Geldart 1986; Kunii & Levenspiel 1991). Agreement with various correlations has tended to be poor when conditions have deviated from those for which the correlation was derived.

Eruption of bubbles or voids at the bed surface causes the initial ejection of particles into the freeboard and the gas flow pattern in the freeboard then determines the distribution of solids concentration and flux. The origin of the entrained particles, whether from the bubble nose or wake, remains a controversial issue. Various mechanistic models for prediction of entrainment rate were reconsidered by Pemberton & Davidson (1986a). Ejection from the bubble roof was found to be predominant for group B particles with $U/U_{mf} < 10$ to 15, while wake ejection plays a greater role for group A, and for group B particles with $U/U_{mf} > 10$ to 15. Pemberton & Davidson (1986b) proposed a three-phase model within the freeboard, with horizontal turbulent diffusion included to describe migration of fine particles towards the outer walls. The lateral mass transfer coefficient for these fine particles is related to turbulence in the freeboard which is believed to originate from puffs of gas (called 'ghost bubbles') caused by bubbles erupting at the bed surface. The model enabled reasonable prediction of the upward flux of the fine particles, the particle concentration and the TDH.

Physical models for entrained particle flux prediction above the TDH have been proposed by Briens & Bergougnou (1986) and Briens *et al.* (1988). These models assume that the particle flux is limited by the maximum carrying capacity and by the flux at the bed surface. The flux caused by ejection at the bed surface was assumed to be strongly influenced by the wakes of bubbles coalescing near the bed surface. Sciazko *et al.* (1991) discussed the interdependence of elutriation rate constant, saturation carrying capacity and choking parameters.

Fung & Hamdullahpur (1993a and 1993b) developed an entrainment flux model based on particle trajectories in the freeboard with consideration of interactions between the dense and lean phases, rather similar in some respects to the approach of Choi *et al.* (1989). Features including bubble eruptions (single or coalescing), bubble-induced gas puffs and the particle ejection mechanism were incorporated in the model. Entrainment is therefore closely related to the bed hydrodynamics. For example, the amount of solids ejected into the freeboard is related to the coalescence frequency, the bubble velocity and the bubble size. Gas velocity fluctuations at the surface may also be influenced by the bubble throughflow velocity (Yule & Glicksman 1989; Levy & Kocatulum 1990). The particle ejection velocity at the bed surface is usually linked to the bubble velocity due to limited experimental data. Good agreement was obtained between model predictions and experimental data on mean gas velocity profiles in the freeboard. Although the predicted particle velocities in the freeboard could explain experimental trends, predictions were significantly larger than corresponding experimental values. More work is required to improve the predictive mechanistic models and to make detailed hydrodynamic measurements in the freeboard region.

Methods of reducing entrainment by increasing the relative humidity and using fine particles have also been investigated (Geldart & Wong 1987; Baron *et al.* 1992). A decrease in entrainment rate was attributed to decreasing electrostatic effects and improved cohesivity. Baron *et al.* (1992) found that humidity did not affect the size distribution of elutriated particles, but it greatly affected the flux. A novel technique employing a layer of floating balls to reduce entrainment was explored by Baron *et al.* (1988b and 1989) and was found to cause a reduction of about 50% in the entrained solids flux above the TDH. In other work, vertical tubes were found to reduce the effective TDH, but the overall carryover above the TDH was unaffected (Pemberton & Davidson 1986b). However, there appeared to be an optimum configuration for the vertical tubes. Further investigation in this area is also warranted.

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7. TURBULENT FLUIDIZATION

While there has been considerable study of the transition to the turbulent fluidization (see section 2.3), there has been remarkably little investigation of the hydrodynamics of the regime itself. The pioneering paper by Abed (1984) remains the most helpful source of information on flow patterns within beds operated in the turbulent regime for type A powders. Horio *et al.* (1992) determined local voidages on the axis of a 50 mm diameter volume with FCC and fine sand. They reported cluster lengths and voidages for both the turbulent and fast fluidization regimes. Clusters existed in both regimes and had internal voidages which did not vary substantially with gas velocity, 0.91 and 0.78 for the FCC and sand, respectively. The clusters were, however, substantially larger in the turbulent regime (where they were typically 20 mm in length) than in the fast fluidization regime (where they were typically 20 mm in length). This led the authors to declare that the turbulent and fast fluidization regimes are intrinsically different regimes, with clusters suspended by the gas flow in the latter but not in the former. Notwithstanding this finding, Weinstein *et al.* (1989) found similar gas axial and radial mixing behaviour in the two regimes.

It is not yet clear whether the turbulent regime constitutes a separate hydrodynamic regime for group B and D solids or simply a transition range of gas velocity where there is a gradual decrease in slug-like behaviour and a corresponding increase in the duration of intermittent periods of fast-fluidization-like behaviour, as suggested by Brereton & Grace (1992). There is clearly a need for more fundamental work on the nature and hydrodynamic properties of the turbulent regime for a range of different particles and operating equipment.

8. CIRCULATING FLUIDIZED BEDS

The operation of high velocity fluidized beds dates back to the early 1940s when FCC technology was first developed (Jahnig *et al.* 1980; Squires 1986; Avidan *et al.* 1990; Grace 1990). However, lower catalyst selectivity and other technical difficulties led to a decrease in operating velocities. It was not until the 1970s that the high velocity fluidized bed was 're-invented' (Reh 1971; Yerushalmi *et al.* 1976).

In the last ten years, major efforts have been devoted to the study of the fast fluidization regime and the development of circulating fluidized bed (CFB) reactors. This can be seen from the increasing share of CFB related papers in the last four international fluidized bed conferences (Kunii & Toei 1984; Østergaard & Sørensen 1986; Grace *et al.* 1989; Potter & Nicklin 1992), in addition to the more than 200 papers compiled in four international conferences on circulating fluidized bed technology (Basu 1986; Basu & Large 1988; Basu *et al.* 1991; Avidan 1994). A multi-authored book on circulating fluidized beds will be published soon (Grace *et al.* 1996). In addition to applications developed prior to 1986 such as fluid catalytic cracking (FCC) (Avidan *et al.* 1990), coal combustion (Kullendorff & Andersson 1986), calcination (Reh 1971) and the Fischer-Tropsch process (Shingles & McDonald 1988; Steynberg *et al.* 1991), CFB has now also been used for direct oxidation of butane to maleic anhydride (Contractor 1988), burning a variety of fuels other than coal (Grace *et al.* 1989; Brereton *et al.* 1991; Itoh *et al.* 1991), gasification (Blackadder *et al.* 1986) and pyrolysis (Berg *et al.* 1989). More than 60 laboratory-scale CFB units have been built around the world (Zhu & Bi 1994).

With the expanded interest in high-velocity systems have also come new developments of CFB reactors such as the cocurrent downflow CFB (downer) (Zhu *et al.* 1995; Zhu & Wei 1995), internal CFB (Milne *et al.* 1992), high density CFB (Bi & Zhu 1993) and, more recently, liquid-solids and three-phase CFBs (Liang *et al.* 1994a and 1994b).

To achieve fast fluidization, solids must be continuously fed to maintain the required solids holdup in a vertical riser. This is usually realized by capturing solids leaving at the top and returning them to the bottom of the riser through a recirculation system. To maintain the fast fluidization regime, the superficial gas velocity must be high enough in the riser that significant numbers of particles can be entrained upwards. Solid particles must be fed into the bed continuously to compensate for material loss due to entrainment. A typical CFB set-up is shown in figure 4. It consists of a riser, where fast fluidization is achieved and where most reactions happen, a gas-solids separator (e.g. a cyclone) to capture the entrained solids, a solids recirculation system (e.g. a



Figure 4. Typical setup of circulating fluidized bed (Yang 1994).

standpipe), and a solids feed system (e.g. L-valve or slide valve). The solids circulation rate is usually controlled by the valve but can be regulated at other places in the loop (e.g. Li & Kwauk 1980).

Compared with conventional low velocity bubbling and turbulent fluidized bed reactors, CFB reactors offer several common advantages such as favourable gas-solids contacting efficiency due to high slip between gas and solids, a more uniform distribution of solids due to reduced gas by-passing, reduced axial gas and solids backmixing, higher gas throughput, independent gas and solids retention time control, improved turndown and possible separate gaseous reactant zones. These advantages are achieved at the expense of some reduction in heat transfer coefficients between heat transfer surfaces and suspension and somewhat greater temperature gradients than in dense beds.

The above advantages make CFB reactor extremely useful for the following:

- (a) reactions of short contact time, e.g. where the intermediate is the desired product,
- (b) reactions with rapidly deactivating catalyst, given the must reduced solids backmixing and ease of solids transportation,
- (c) reactions requiring considerable addition or release of heat where solids can be used as heat carriers,
- (d) gas-solids reactions where high gas-particle contact efficiency is essential,
- (e) processes with varying feed and product requirements, given the flexibility of operation.

Since the key features of CFB result from the gas-solids co-current upflow in the riser where most reactions occur, this section concentrates on the riser. Other components, such as gas-solids separators and the solids return and feed systems are not considered, except to the extent needed to understand behaviour in the riser.

8.1. Axial flow structure

The axial variation of cross-sectional average voidage in the riser was first studied by Li & Kwauk (1980), who reported on S-shape profile with a dense region at the bottom and a dilute region at the top of the riser. This profile has since been observed by others (Hartge *et al.* 1986a; Schnitzlein & Weinstein 1988) and for some time was considered universal. In recent years, however, other profiles have also been observed (Arena *et al.* 1988; Hartge *et al.* 1988; Horio *et al.* 1988; Li *et al.* 1988a; Bai *et al.* 1992a; Hirama *et al.* 1992b; Wong *et al.* 1992; Brereton & Grace 1994; Harris

et al. 1994). The bottom dense region may not always be present and a denser region may exist at the top for constricted exits. Operating variables such as solids circulation rate (Arena et al. 1988; Li et al. 1988a; Hirama et al. 1992b), total solids inventory (Li et al. 1988a; Bai et al. 1992a), particle size and density (Bai et al. 1992a), solids inlet configuration (Bai et al. 1992a), riser exit structure (Jin et al. 1988; Bai et al. 1992a; Martin et al. 1992; Brereton & Grace 1994; Harris et al. 1994), secondary air injection (Lasch et al. 1988; Wang & Gibbs 1991; Arena et al. 1993; Brereton & Grace 1994) and the level of solids reintroduction into the riser (Brereton & Grace 1994), affect the axial voidage profile.

Li et al. (1988a) proposed that the shape of the axial voidage profile depends on the relative magnitude of the solids circulation rate to the saturation solids carrying capacity of the gas. An exponential shape (Bai et al. 1992a) can be observed for circulation rates less than the saturation carrying capacity, while an S-shape or an almost straight line profile occurs for circulation rates greater than the saturation carrying capacity, as shown in figure 5. Profiles were classified by Yang (1988) into three types, a dilute phase transport regime (type I, exponential shape), the fast fluidization regime (type II, S-shape) and the dense transport regime (type III, straight line), all of which can be achieved in a circulating fluidized bed.

Bai et al. (1992a) conducted systematic experiments to show how operating conditions affect the axial voidage profile. As summarized in figure 6, they found that

- (a) decreasing superficial gas velocity and increasing solids circulation rate increase the solids holdup, especially in the bottom region,
- (b) increasing particle size or particle density has a mixed effect, resulting in slightly less solids holdup in the upper region, but much higher solids holdup in the bottom region where it may form a rather dense bed,
- (c) decreasing riser diameter leads to a reduction in bed voidage, especially in the bottom region (see also Arena *et al.* 1988).

These trends are consistent with other reported data (Hirama et al. 1992b; Arena et al. 1988; Li et al. 1988a). For all cases, a dense bed may appear at the bottom at conditions favouring high



Figure 5. Classification of types of axial bed voidage profiles observed in the riser of circulating fluidized beds (modified from Yang 1988).



Figure 6. Illustration of the various factors influencing average voidage distribution: the arrows indicate effect of: (A) decreasing gas velocity; (B) increasing solids circulation rate, (C) decreasing particle diameter, (D) decreasing particle density, (E) decreasing column diameter, (F) decreasing exit restriction, (G) influence of bed height with different exit restrictions, (H) decreasing inlet restriction, (I) increasing solids inventory with strong inlet restriction and (J) increasing solids inventory with weak inlet restriction (from Bai et al. 1992a).

solids holdup. For example, Arena *et al.* (1988) showed that increasing solids circulation rate at a fixed superficial gas velocity leads to the formation of a dense phase region at the bottom, its height increasing with solids circulation rate.

The exponential shape occurs when solids entering the riser are immediately entrained so that there is no significant particle accumulation at the riser bottom. A strong restrictive riser exit (such as a right angle bend) increases the solids holdup in the top region of the riser due to particle rebounding (Jin *et al.* 1988; Hirama *et al.* 1992a; Martin *et al.* 1991; Brereton & Grace 1994; Harris *et al.* 1994; Zheng & Zhang 1994; Zhou *et al.* 1994). By utilizing orifices of different sizes at the riser exit, Harris *et al.* (1994) demonstrated that a stronger exit restriction (smaller orifice diameter) significantly increases solids holdup in the top region. A C-shaped profile is found for a constricted exit (Martin *et al.* 1992), but not for a weak exit restriction such as a smooth bend at the riser top (Bai *et al.* 1992a; Brereton & Grace 1994).

When the total solids inventory in the system is increased and the solids inlet restriction is reduced, a significant dense region may form at the bottom of the riser, leading to an S-shaped axial voidage profile (Arena *et al.* 1988; Bai *et al.* 1992a; Hirama *et al.* 1992a; Mori *et al.* 1992). If the exit is restricted sufficiently, the S-shaped profile is superimposed by a C-shaped profile in

the top region (Bai *et al.* 1992a; Wong *et al.* 1992). The existence and the length of the bottom dense phase region result from the system pressure balance (Rhodes & Geldart 1986b; Li *et al.* 1988a; Yang 1988; Bai *et al.* 1992a; Bi & Zhu 1993). The pressure at the riser bottom must equal the pressure at the bottom of the return column minus the pressure drop across the solids recycle valve. If the pressure at the bottom of the return column is increased due to increased solids inventory or the pressure drop across the feeding device is reduced due to a reduced inlet restriction, solids holdup in the riser must also increase (Bi & Zhu 1993).

The above profiles can be varied by the axial position of solids feeding and by injection of secondary air. Feeding solids well above the gas distributor can cause a localized high solids holdup in the vicinity of the solids feed position (Brereton & Grace 1994). The effects of secondary air injection have been studied in relation to CFB combustion (Wang & Gibbs 1991; Brereton & Grace 1994). In all cases (Ilias *et al.* 1988; Lasch *et al.* 1988; Wang *et al.* 1991; Arena *et al.* 1993; Brereton & Grace 1994), keeping the total gas flowrate unchanged, radially injecting a portion of the gas flow at a higher position increases the bed density in the lower region. Tangential secondary air injection increases the bed density in the vicinity of the injection point as well, since the swirling gas flow entrains downflowing solids in the wall region, leading to substantial solids buildup in a swirling wall layer (Ilias *et al.* 1988; Wang & Gibbs 1991; Brereton & Grace 1994). When the tangential velocity is very high, this solids buildup can extend all the way to the top of a short riser (Lasch *et al.* 1988).

Differential pressure measurements have often been used to estimate suspension densities, based on the assumption that pressure losses due to particle-wall friction and particle acceleration are negligible. Comparing directly measured solids holdup along the riser by using a series of quick-closing valves with the values inferred from differential pressure, Arena *et al.* (1986) found that the values calculated from differential pressure measurements are significantly higher in the transition region where particles accelerate very quickly. This discrepancy can result in a change of axial voidage profile from an exponential shape to an apparent S-shape under certain operating conditions (Arena *et al.* 1986). Taking the extra pressure drop due to solids acceleration into account, Weinstein & Li (1989) evaluated the actual solids holdup from differential pressure measurements. Louge & Chang (1990) developed a model which attributes the voidage discrepancies to rapid vertical variations during the transition from dense phase to dilute phase flow.

Upon entering the riser bottom, solid particles usually travel horizontally or downwards due to horizontal or downwards inclined solids feeding. Arena *et al.* (1986) and Bai *et al.* (1990) considered the acceleration zone as extending from the riser bottom to a point at which the pressure gradient becomes constant, while Weinstein & Li (1989) used the criterion of particles attaining a constant velocity.

For risers with a dense phase region at the bottom, it seems appropriate to consider the particle acceleration zone starting at the upper end of the dense phase region since the particle velocity in the dense phase is still very low compared with that in the upper dilute region. This is supported by the dramatic change of axial bed voidage across the transition region between the dense and dilute regions. On the other hand, particles can also be considered to be accelerated in the region immediately above the distributor. This section can be considered as the initial particle acceleration zone but, given the low particle velocities in the dense phase region, does not contribute significantly to overall particle acceleration. Locations of particle acceleration zones under different conditions are shown in the axial voidage profiles in figure 6, which has been modified from the original work of Yang (1988) to reflect the above argument.

Bai *et al.* (1990) pointed out that the acceleration length often occupies $\frac{1}{3}$ to $\frac{2}{3}$ of the riser so that this section cannot be neglected in modelling and reactor design. For practical purposes, however, it is probably sufficient to consider only the rapid acceleration section where most (say 90% to 95%) of the acceleration has been achieved. With this approximate definition, the data presented by Arena *et al.* (1986) and Bai *et al.* (1990) suggest that the acceleration zone coincides with the transition region. This length has been shown by Bai *et al.* (1990) to increase with solids circulation rate and riser diameter, while decreasing with superficial gas velocity. Above the transition region or the solids acceleration region, the cross-sectionally averaged particle velocity experiences little change until near the top, where particles may decelerate with a strong exit restriction (Jin *et al.* 1988; Bai *et al.* 1992a; Brereton & Grace 1994).

Internals are sometimes introduced to improve gas-solids contact in the riser (Davies & Graham 1988; Gan et al. 1990; Jiang et al. 1991; Zheng et al. 1992a; van der Ham et al. 1993). Davies & Graham (1988) showed that swages evenly spaced along a circular riser wall cause the pressure drop in the fully developed region to be lower than for a bare column. On the other hand, Jiang et al. (1991) observed an increase in pressure drop when four ring baffles of open area 56% were installed in a 0.1 m riser, suggesting an increase in solids holdup, although, due to the extremely complex flow behaviour associated with the baffles, it is questionable whether bed voidage can be accurately obtained from the axial pressure profile. For a novel ring-shaped internal, Zheng et al. (1992a) measured a reduced cross-section solids concentration with an optical fibre probe, believed to be due to scraping of superfluous particles from the wall. Filling the riser with regularly packed bars, van der Ham et al. (1993) measured 1.6 to 5 times higher solids holdup compared with an empty column. Although part of this increase was due to particles settling on top of the bars, at least some was due to the effect of the internals on the flow structure. It would appear that baffles with less flow restriction (i.e. little reduction in cross-sectional area) mainly increase the actual gas velocity by displacing downward-moving solids from the wall, leading to higher bed voidage as observed by Davies & Graham (1988) and Zheng et al. (1992a), while baffles with a significant reduction of cross-sectional area obstruct the solids flow, leading to lower bed voidage (Jiang et al. 1991; van der Ham et al. (1993).

8.2. Radial flow structure

Using an isokinetic sampling probe in a riser of diameter 300 mm, van Breugel *et al.* (1970) reported a parabolic radial solids flux profile. Gajdos & Bierl (1978) and Qin & Liu (1982) appear to have been the first to present radial voidage profiles showing high voidage in the centre and much lower voidage close to the wall. This non-uniformity has been confirmed by many others. A core-annulus flow structure was proposed by Bierl & Gajdos (1982) and adopted by others (e.g. Weinstein *et al.* 1984, 1986a and 1986b; Dry 1986). The riser is assumed to consist of a dilute core region, occupying most of the cross-section, and a dense annular region adjacent to the wall where solids mainly travel downwards.

Typical radial voidage profiles are shown in figure 7. A dilute core and denser annulus bed structure are observed. Bed voidage is seen to be high and relatively uniform in the central region extending out to about 70 to 85% of the column radius, after which solids concentration increases dramatically towards the wall, even approaching the solids bulk density in some cases (Hartge *et al.* 1988).

The exact radial solids concentration distribution depends on the operating conditions. Zhang *et al.* (1991) showed that decreasing the superficial gas velocity or increasing the solids circulation rate increases the overall solids concentration and steepens the profile (figure 7). (See also Weinstein *et al.* 1986a; Kato *et al.* 1991; Brereton & Grace 1993; Zhou *et al.* 1994.) For the same superficial



Figure 7. Influence of operating conditions on the radial voidage distribution in the dilute region of the riser (Zhang *et al.* 1991) (D = 0.09 m, H = 10 m, $d_p = 34-72 \ \mu$ m, $\rho_p = 607-2000 \ \text{kg/m}^3$) (a) $U_G = 2.62 \ \text{m/s}$, G_s in kg/m²s; (b) $G_s = 43 \ \text{kg/m}^2$ s; U_G in m/s.

gas velocity and solids circulation rate, the profile becomes more uniform with increasing height (Kato et al. 1991; Yang et al. 1991a; Brereton & Grace 1993).

Herb *et al.* (1989) suggested that there may be a universal radial profile of bed voidage ε for any time-averaged solids concentration. This was supported by Tung *et al.* (1988, 1989) and Zhang *et al.* (1991) for different particles at different superficial gas velocities and solids circulation rates in different risers. The radial distribution can therefore be considered to be a function only of the average cross-section bed voidage $\overline{\varepsilon}$. Tung *et al.* (1989) and Zhang *et al.* (1991) suggested:

$$\varepsilon = \overline{\varepsilon}^{0.191 + (r/R)^{2.5} + 3(r/R)^{11}}.$$
[12]

Using a gamma-ray technique, Azzi *et al.* (1991) obtained three-dimensional radial solids concentration distributions in a laboratory-scale CFB and in an industrial riser, showing that the solids distribution in the tangential direction is also not always uniform, even in the fully developed region. Zhou *et al.* (1994) showed that the lateral voidage distribution in a column of square cross-section is not always symmetrical, especially in the bottom and top regions where particles are fed and withdrawn from one side of the riser. They also found some 'M-shaped' voidage profiles with the minimum solids concentration located well away from the riser axis, a finding consistent with data from several earlier studies obtained in columns of circular cross-section (Weinstein *et al.* 1986b; Hartge *et al.* 1988; Bai *et al.* 1991a; Kato *et al.* 1991; Herb *et al.* 1992).

Figure 8 presents results of Zhou *et al.* (1994), showing that solids holdup in the corners is higher than elsewhere along a flat wall. Membrane walls create significant local effects, increasing the local solids holdups in the shielded fin region (Lockhart *et al.* 1995).

Isokinetic and non-isokinetic sampling probes have been used to measure the radial distribution of solids flux in the vertical direction (van Breugel *et al.* 1970; Monceaux *et al.* 1986a and 1986b; Bader *et al.* 1988; Rhodes *et al.* 1988; Rhodes 1990; Rhodes & Laussmann 1992; Herb *et al.* 1992; Bodelin *et al.* 1994). In principle, isokinetic sampling is superior to non-isokinetic sampling. However, given the relatively large particles, there is normally little need for isokinetic sampling for CFBs. In any case, none of the isokinetic sampling probes reported so far has the ability to follow the instantaneous changes in local gas velocity.

Both upwards and downwards solids fluxes were measured by Herb *et al.* (1992). As shown in figure 9, the central core region is dominated by an upflowing solids suspension flux. The presence of a small downward solids flux in the core suggests occasional downwards-moving particle clusters there. The annulus, with a thickness an order of magnitude smaller than the column width or diameter, is occupied by a dense, downflowing solids suspension. Similar profiles have been reported by others (Bader *et al.* 1988; Rhodes *et al.* 1988 and 1989).

Monceaux *et al.* (1986b) and Herb *et al.* (1992) showed that increasing solids circulation rates lead to steeper profiles, with higher upflow flux in the core and a greater downflow in the annulus. Increasing superficial gas velocity, on the other hand, gives an opposite effect. Herb *et al.* (1992) reported that there was no significant difference in dimensionless profiles between two risers of diameter 0.05 and 0.15 m. Both Rhodes (1990) and Wei *et al.* (1994a) found that the radial profile becomes more uniform with height, contrary to Herb *et al.* (1992). However, the conclusion of the latter group that the radial solids flux profile becomes less uniform up the riser was based on only two axial locations, the lower of which may have been in the dense phase region. Rhodes & Laussmann (1992) measured the tangential variation of solids flux as well as the radial distribution in a 0.152 m riser. The solids flux varied significantly with angular position, especially in the annular region.

The radial solids flux was measured by Qi & Farag (1993). Their results indicate that the flux in the radial direction is small in the central core region but increases quickly towards the wall, with radial flux reaching values exceeding the solids circulation rate. Such high lateral fluxes have not been confirmed in recent work (Zhou *et al.* 1995b). Qi & Farag (1993) also measured the radial distribution of solids holdup and concluded that the radial solids flux is proportional to the local solids holdup. Given the role of lateral flux in determining heat transfer, further work is clearly required in different columns.

Fewer studies have been conducted on the radial distribution of gas velocity, no doubt due to difficulties in measurement. Using a fibre optic probe laser-Doppler velocimeter with 1 μ m talcum particles as tracers, Yang *et al.* (1993) reported results shown in figure 10. A clear core-annulus



Figure 8. Lateral bed voidage profiles in the dilute region of a 146 × 146 mm square riser, $U_G = 7.0$ m/s, $G_s = 40$ kg/m²s, Ottawa sand ($d_p = 213 \ \mu m$, $\rho_p = 2640$ kg/m³) (from Zhou *et al.* 1994).

flow structure is again evident. The local gas velocity is high and does not change significantly in the central core region, while the local gas velocity is lower with a much steeper profile in the thin annulus region. Figure 10 shows that an increase in solids circulation rate leads to an increase in local gas velocity in the central core region and a decrease near the wall. A possible explanation (Yang *et al.* 1993) is that the solids concentration in the near-wall region increases sharply with increasing solids circulation rate, creating more resistance to gas flow in the annular region. For the same solids circulation rate, increasing the superficial gas velocity leads to a more uniform distribution of local gas velocity, a trend similar to the radial distributions of bed voidage and solids flux discussed above.

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All observed radial distribution profiles of particle velocity support the core-annulus flow structure where solids are mainly entrained upwards in the core region and descend in the annular region (Bader *et al.* 1988; Horio *et al.* 1988; Yang *et al.* 1990; Yang *et al.* 1991a, 1991b and 1991c; Harris & Davidson 1992; Yang *et al.* 1993; Bouillard & Miller 1994; Harris *et al.* 1994; Zhou *et al.* 1995a). Typical radial distributions of particle velocity (both upwards and downwards) obtained by Bader *et al.* (1988) are shown in figure 11. Profiles of average net particle velocity have been reported by Horio *et al.* (1988), Yang *et al.* (1991a and 1991b) and Harris & Davidson (1992). Note that although the upflow and downflow particle velocities are similar in magnitude near the wall in figure 11, the downflow solids flux is much higher (see figure 9) since there are many more particles travelling downwards there.

Dry (1987) found that at superficial gas velocities of 6 to 9 m/s, segregation of particles occurs in the radial direction, with a preponderance of coarse particles in the annulus region. This segregation was not observed at a superficial gas velocity of 2 m/s. Bodelin *et al.* (1994) obtained similar results and suggested that this is due to a high internal recirculation rate of coarse particles. Further studies are necessary to confirm these findings.

Radial particle migration from the dilute core towards the higher concentration wall region has been attributed to turbulent diffusion (Bolton & Davidson 1988) and to the saturation of gas



Figure 10. Radial distribution of gas velocity in the dilute region of a CFB riser of diameter 0.14 m and overall height 11 m at a height of 3.2 m (FCC particles $d_p = 54 \,\mu$ m, $\rho_p = 1550 \,\text{kg/m}^3$) (from Yang *et al.* 1993).



Figure 11. Radial distribution of upwards and downwards particle velocity in the dilute region of a CFB riser of diameter 0.305 m and height 12.2 m, $U_G = 3.7$ m/s, $G_s = 98$ kg/m²s, FCC particles ($d_p = 76 \mu$ m, $\rho_p = 1714$ kg/m³) (from Bader *et al.* 1988).

carrying capacity in the core region (Yang 1988). Radial particle migration is also predicted by several hydrodynamic models (Gidaspow *et al.* 1989; Sinclair & Jackson 1989; Tsuo & Gidaspow 1990; Senior & Brereton 1992; Wu 1994; Yasuna *et al.* 1994).

The hydrodynamics in the bottom dense phase region is also of interest. Few studies have been reported. The lower region has been considered a turbulent fluidized bed by some researchers (Schnitzlein & Weinstein 1988; Brereton & Grace 1992) and a bubbling bed by others (Bolton & Davidson 1988; Johnsson *et al.* 1992; Svensson *et al.* 1993). Werther (1994) suggested that the lower bed may be turbulent in a laboratory riser but bubbling in a larger scale riser. Group A powders are more likely to lead to turbulent fluidization, while group B particles are more prone to bubbling fluidization. However, the bottom region cannot always simply be treated as either a bubbling bed or a turbulent bed, given the high superficial gas velocities in CFB units, unless there is secondary air introduction, below which the superficial gas velocity is considerably reduced. There is evidence of strong radial voidage non-uniformity in the lower region (Hartge *et al.* 1986a; Schnitzlein & Weinstein 1988), exceeding that in 'conventional' turbulent fluidized beds (Abed 1984).

To break up the radial flow structure discussed above, internals can be installed to redistribute gas and particles (Davies & Graham 1988; Gan *et al.* 1990; Jiang *et al.* 1991; van der Ham *et al.* 1993). Gan *et al.* (1990) found that a bluff body (half oval with parabolic curvature, flat end up, with its large upper end occupying 50% of the riser cross-section) on the axis of a riser significantly changed the radial gas and particle flow structures downstream. Gas and solids contact were found to be greatly improved due to the breakup of the non-uniform radial gas and particle distribution by the bluff body. Regularly packed bars were observed to have a similar effect, effectively suppressing radial inhomogeneities and thereby greatly improving gas-solids contact (van der Ham *et al.* 1993).

8.3. Micro flow structure

In early work on CFB hydrodynamics, Yerushalmi *et al.* (1976) and Li & Kwauk (1980) reported that particles tend to exist in aggregated forms. A theoretical analysis was also proposed (Grace & Tuot 1979) to explain the origin of particle clusters.

Time variations of gas and solids flow have been recorded by many researchers. For example, the change of local solids concentration with time has been monitored using optical fibre probes (Hartge *et al.* 1988; Jiang *et al.* 1994) and capacitance probes (Herb *et al.* 1989; Brereton & Grace

1993). Several methods have been used to characterize this time variation of solids concentration, including standard deviations of local particle concentration (Brereton & Grace 1993), probability density functions (Hartge *et al.* 1988; Herb *et al.* 1989), an intermittency index (Brereton & Grace 1993; Zhou *et al.* 1994) and an intermittency factor (Jiang *et al.* 1994).

The intermittency index, defined by Brereton & Grace (1993) by

Intermittency index

= Standard deviation of density fluctuations at a given point [13] Standard deviation of density fluctuations for fully segregated two-phase flow with identical time-mean density at the same point

is constrained between zero, when there is a uniform local suspension, and unity, when regions of gas-alone alternate with clusters of voidage equal to the loose-packed voidage. A higher value indicates a more segregated and time-varying flow structure at that location. The radial distribution of the intermittency index is shown in figure 12. It is shown that the index varies from 0.1 to 0.6, indicating that the two-phase flow is neither uniform nor completely segregated. Except for one set of data from the dense phase or transition region, the index increases from the core to the annulus, suggesting that the flow is more uniform in the core than in the annular wall region. The index is around 0.1 to 0.2 in the core region so that the flow structure is rather uniform there. This intermittency index has also been determined by Zhou *et al.* (1994) in a square column where similar trends were observed. The index was found to be slightly higher in a corner than elsewhere along the wall. The intermittency index at the axis and the wall both decreased gradually with height, indicating that the flow becomes somewhat more uniform.

The micro-flow structure was observed by Bai *et al.* (1991c) employing a two-dimensional column, by Li *et al.* (1991a and 1991b) using a video camera and a special optical fibre image probe, and by Horio & Kuroki (1994) and Kuroki & Horio (1994) using a laser sheet technique, Li *et al.* (1991a and 1991b) show that there are two distinct phases, a dispersed phase, in which solid particles are essentially present individually, and a cluster phase. This is consistent with the photos of Kuroki & Horio (1994) who observed that clusters exist even for very dilute conditions. Lateral transportation of clusters was observed by all three groups of researchers. From very localized micro-visualization, Li *et al.* (1991a and 1991b) reported that clusters have rather irregular shapes and highly variable size. The clusters were observed to transform from strands at the centre of the column into near-spheres (swarms, according to the classification presented below) adjacent to the wall. On the other hand, Bai *et al.* (1991c), Horio & Kuroki (1994) and Kuroki & Horio (1994) observed that most clusters have a U-shape with a round nose facing downward and with particles shed continuously from the periphery.

Wei *et al.* (1994a and 1994c) employed an optical fibre image system to observe the gas-solids suspension and found significant axial and radial variations. From the riser centre to the wall, both cluster size and frequency increased. Increasing superficial gas velocity and/or decreasing solids circulation rate reduced the cluster size and frequency of appearance. Cluster size decreased with height.



Figure 12. Radial variations of intermittency index, $U_G = 6.5 \text{ m/s}$, $G_s = 48 \text{ kg/m}^2\text{s}$, sand particles $(d_p = 148 \ \mu\text{m}, \ \rho_p = 2650 \text{ kg/m}^3)$ for a column of diameter 0.152 m and height 9.3 m (from Brereton & Grace 1993).

Forms of congregation	Region	Shape	Character	Scale	Origin
Particle clusters	core and annulus	various	$\frac{D_{\rm e} < 1 \rm cm}{\varepsilon \approx \varepsilon_{\rm mf}}$	micro-scale	 (1) interparticle forces (2) particle-wake interaction
Particle streamers /strands	core and annulus	bands	$L_{\rm e} > 1 {\rm cm}$ $\varepsilon \approx 0.7 - 0.95$ $U_{\rm e} > 0$	meso-scale	 resemble clusters breakup of particle sheets non-uniform introduction of particles
Particle swarms	annulus	approx. spherical	$L \approx 1-1.5 \text{ cm}$ $U_{e} \approx -0.3 \sim$ -0.4 m/s	meso-scale	(1) wall-particle interaction
Particle sheet	annulus	layer	$\delta_{\rm e} \approx 0-3 {\rm cm}$ $U_{\rm e} \approx -1 {\rm m/s}$	macro-scale	 (1) resemble particle swarms (2) particles diffusing to the wall region and swarms

Table 1. Summary of solids congregation forms in CFBs (Bi et al. 1993a)

 D_e = equivalent size, L_e = equivalent length, U_e = average velocity, ε = average voidage, δ_e = average thickness of particle sheets.

There are many definitions for clusters (Yerushalmi et al. 1976; Grace & Tuot 1979; Li & Kwauk 1980). It is convenient to define clusters simply as dense clouds of particles having significantly more particles per unit volume than the surrounding dilute regions (Bi et al. 1993a). Many terms have been used (sometimes interchangeably) to describe these clusters. The terms 'streamers', 'strands', 'ribbons' and 'dense packets' have often been used interchangeably. In an effort to classify the different types of particle aggregations, Bi et al. (1993a) proposed four categories summarized in table 1:

- (a) Particle clusters: Groups of several to dozens of particles congregated in the riser in order to reduce the effective drag force exerted on them. The shape of these assemblies may be in the form of strings, triangles or other shapes.
- (b) Particle streamers/strands: Dense bands of larger size than clusters observed in the core region and along the walls of circulating fluidized beds (e.g. Yerushalmi *et al.* 1976; Li *et al.* 1991a and 1991b; Takeuchi & Hirama 1991; Horio *et al.* 1992).
- (c) Particle swarms: Relatively dense particle assemblies at the bed wall, especially when a riser is operated under relatively dilute conditions, as observed by Bai *et al.* (1991b) and Rhodes *et al.* (1992a). They are generally approximately spherical (Li *et al.* 1991a and 1991b).
- (d) Particle sheets: Two-dimensional layers of densely populated particles falling near the bed wall. These are believed to result from lateral particle segregation (Hartge *et al.* 1988; Bolton & Davidson 1988; Senior & Brereton 1992).

Particle clusters and particle streamers may originate due to instability of gas-solids transport (Grace & Tuot 1979), energy minimization of gas-solids transport (Li *et al.* 1988b) or particle-wake interaction (Zenz & Othmer 1960; Fujima *et al.* 1988 and 1991). Both stability and energy considerations suggest that particles tend towards congregated forms rather than remaining dispersed homogeneously. According to the wake theory, particles are drawn into the low-pressure wake region behind other particles (Bai *et al.* 1991b), where they experience reduced drag. Individual particles experience catching, touching, collisions and separation. The behaviour of particle clusters and streamers in the core region can also be successfully described by this theory (Fujima *et al.* 1991).

Particle sheets and swarms descending along the wall are retarded by friction at the column wall. Addition of more particles from above, on the other hand, speeds their descent. Eventually a balance is reached with particle swarms growing to an equilibrium size. The inside surface becomes wavy due to instabilities as in two-phase gas-liquid flows. 'Satellites' are discharged intermittently from the wavy peaks as continuous addition of particles occurs from above (Bi *et al.* 1993a). The size of particle clusters was observed by Li *et al.* (1991a and 1991b) using image probes to be of the order of several millimeters for FCC particles, similar to the size deduced from the local slip velocities (Yang *et al.* 1993). Cluster voidages have often been considered to be equal to ε_{mf} (e.g. Yerushalmi *et al.* 1978) or ε_{mb} (Bai *et al.* 1989), but in reality there is a range of voidages (Wu *et al.* 1991).

Particle streamers or strands have also been observed in the core of the riser (Yerushalmi *et al.* 1976; Li *et al.* 1991a and 1991b; Horio *et al.* 1992; Kuroki & Horio 1994). Horio *et al.* (1992) fixed an optical fibre probe at the axis of a riser to deduce the voidage and length of particle streamers. Their voidages were found to range from 0.7 to 0.95 depending on particle properties, while being almost independent of gas velocity and solids circulation rate. The average streamer length varied from 50 to 5 mm with increasing gas velocity.

Particle sheets have been broadly reported to exist in the wall region of risers, although different names are commonly used in the literature (Hartge *et al.* 1988; Rhodes *et al.* 1989 and 1992a; Horio & Kuroki 1994; Kuroki & Horio 1994). The thickness of these sheets can be considered to be more or less the same as the thickness of the annulus, while the boundary between the core and the annulus can be estimated as the point where the axial time-mean particle velocity is zero (Bi *et al.* 1993a). Based on this definition, the thickness may be up to 20 mm in laboratory-scale risers (Hartge *et al.* 1988; Rhodes *et al.* 1989). The falling velocity of the particle sheet is typically around 1.0 m/s (Hartge *et al.* 1988; Rhodes *et al.* 1992a; Wu *et al.* 1990). Louge *et al.* (1990) found that the average voidage at the wall ranged from 0.45 to 0.6. Wu *et al.* (1991) found a wide probability distribution of wall voidage. Bi *et al.* (1993a) suggested that the bed voidage fluctuations and velocity fluctuations are most significant at the core-annulus boundary region due to the abrupt formation and disintegration of particle sheets. This is supported by voidage fluctuation data (Rhodes *et al.* 1992a; Brereton & Stromberg 1986), gas and solids velocity fluctuation data (Yang *et al.* 1991a and 1993) and slip velocity data (Yang *et al.* 1993).

Particle swarms were studied by Rhodes *et al.* (1992a). Swarms were seen to fall in contact with the column wall with a typical velocity from 0.3 to 0.4 m/s, significantly lower than that of particle sheets. Swarms were observed at low solids circulation rates where particle sheets were rarely identified, in agreement with observations by Bi *et al.* (1993a) in a two-dimensional column. The typical stable chord length of such particle swarms was around 10 to 15 mm (Bi *et al.* 1993a). The voidage of particle swarms can be considered the same as particle sheets discussed above.

The overall relative velocity in circulating fluidized beds, given by

$$\overline{U}_{\rm slip} = \frac{U}{\varepsilon} - \frac{G_{\rm s}}{\rho_{\rm p}(1-\varepsilon)},$$
[14]

has been found to be much higher than the terminal velocity of single particles (Yerushalmi & Cankurt 1979), and this has been attributed to the existence of particle clusters or streamers (Yerushalmi & Cankurt 1979; Grace & Tuot 1979; Li *et al.* 1991a and 1991b). Geldart & Rhodes (1986), on the other hand, suggested that such a high 'apparent' slip may simply be due to lateral segregation discussed above.

Yang et al. (1992 and 1993) appear to be the only authors to report a systematic study of local relative velocity. Local gas and particle velocities were measured simultaneously to obtain a time-average at several radial positions in a 140 mm diameter, 11 m tall laboratory-scale circulating fluidized bed, using a fibre optic probe LDV technique. Solids holdups were measured at the corresponding positions. Typical radial profiles of local relative velocity reported by Yang et al. (1993) are shown in figure 13. Measured local relative velocities are significantly lower than the overall relative velocity calculated from [14]. Yang et al. (1993) concluded that the difference is mainly due to lateral particle segregation, with particle aggregation playing a secondary role. Relative velocity in the central region is seen to be always smaller than near the wall, corresponding to fewer aggregates. A maximum point is reached near the wall or near the boundary between the upward and downward solids flow regions. It is seen that the local relative velocity increases slightly with solids circulation rate and decreases with superficial gas velocity, probably due to the variation of solids concentration. Yang et al. (1993) showed that the radial profiles of relative velocity are related to the magnitude of gas and particle velocity fluctuations. If the measured local relative velocity is treated as the effective terminal velocity of particle clusters, effective particle cluster sizes can be estimated (Yerushalmi & Cankurt 1979).

8.4. Gas and solids mixing in CFB

The downwards motion of particles in the wall region described above causes backmixing of solids and may also lead to downflow (backmixing) of gas in the annulus. This results in non-uniform residence time distributions, which are undesirable for some chemical reactions. On the other hand, radial dispersion of gas and solids is usually beneficial to overall conversion.

Gas mixing has been studied by many researchers. The axial gas flow deviates significantly from plug flow (Bader *et al.* 1988; Brereton *et al.* 1988; Li & Wu 1991; Bai *et al.* 1992b). One-dimensional dispersed plug flow models have been employed by several researchers to correlate experimental data (Bai *et al.* 1992c; Martin *et al.* 1992; Werther *et al.* 1992a). Reported axial gas dispersion coefficients mostly fall in the range of 0.1 to $0.6 \text{ m}^2/\text{s}$ (Bader *et al.* 1988; Li & Wu 1991; Bai *et al.* 1992c; Martin *et al.* 1992b).

Gas backmixing is closely related to the operating conditions (Brereton *et al.* 1988; Dry & White 1989; Li & Wu 1991; Bai *et al.* 1992b and 1992c), increasing with solids circulation rate (Brereton *et al.* 1988; Li & Wu 1991; Bai *et al.* 1992b and 1992c) and decreasing with superficial gas velocity (Li & Wu 1991; Bai *et al.* 1992b). A systematic study was conducted by Bai *et al.* (1992b) to measure residence time distributions, showing larger peak values with shorter tails as the superficial gas velocity is increased, but smaller peak values with longer tails as the solids circulation rate is increased. At a fixed gas velocity, increasing solids circulation, leading to a less uniform radial gas distribution. The increased local gas velocity in the centre and the reduced (or increased downwards) gas velocity at the wall cause more gas to pass through the core region, increasing gas dispersion. The opposite occurs when gas velocity is increased at a fixed solids circulation rate (Bai *et al.* 1992b).

Radial gas dispersion has been measured by several researchers (Yang *et al.* 1984; Adams 1988; Bader *et al.* 1988; Martin *et al.* 1992; Werther *et al.* 1992a; Zheng *et al.* 1992b; Amos *et al.* 1993). Except for the work of Yang *et al.* (1984), where an extremely low dispersion coefficient was reported, all reported dispersion coefficients were of order 0.002 to $0.02 \text{ m}^2/\text{s}$, one to two orders of magnitude less than the axial dispersion coefficient. The influences of operating conditions on the radial gas dispersion appear to be complex. In the core region of the dilute upper part of the riser, Werther *et al.* (1992a) reported that radial gas dispersion is independent of superficial gas velocity and independent of solids circulation rate for solids circulation rates from 0 to 70 kg/m²s. Zheng *et al.* (1992b), on the other hand, suggested that there is a threshold solids circulation rate below which radial gas dispersion increases with superficial gas velocity and above which radial gas dispersion decreases with superficial gas velocity. More studies are needed to provide a clearer picture.



Figure 13. Radial distribution of slip velocity in the dilute region 3.2 m above the distributor of a CFB riser of diameter 0.14 m and overall height 11 m, FCC particles ($d_p = 54 \ \mu m$, $\rho_p = 1550 \ \text{kg/m}^3$) (from Yang *et al.* 1993).

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Axial and radial solids mixing have also been studied (Bader et al. 1988; Bolton & Davidson 1988; Ambler et al. 1990; Milne & Berruti 1991; Bai et al. 1992c; Martin et al. 1992; Rhodes et al. 1992b; Weinell & Dam-Johansen 1992; Mori et al. 1993). Solids backmixing is mainly due to the downflow of particles in the annular region (Bader et al. 1988). There are few data available. From the results of Bai et al. (1992c), axial solids dispersion increases somewhat at high solids circulation rates, but a correlation due to Rhodes et al. (1992b) suggests the opposite. Increasing gas velocity increases the axial solids dispersion, contrary to the influence of gas velocity on axial gas dispersion. Bai et al. (1992c) found that fine particles have a more uniform residence time distribution than coarser particles, since fine particles are more uniformly distributed in the riser than coarser particles.

Bai *et al.* (1992c) and Martin *et al.* (1992) concluded that axial solids mixing is greater than axial gas mixing due to the significant internal solids circulation. Bai *et al.* (1992c) showed that the residence time distributions for gas are relatively narrow with high peak values, while the solids have wider distributions with lower peak values and longer tails, indicating more axial dispersion and backmixing of solids. This difference, however, diminishes as the superficial gas velocity is increased, due to more uniform suspension of solids in the riser (Bai *et al.* 1992c). At high gas velocities, the gas–solids flow patterns approach those of dilute phase transport where the relative velocity tends to be constant and close to the particle terminal velocity.

Little information is available on the radial exchange of particles between the core and the annulus, and no data have been reported on the radial solids dispersion coefficient. Mori *et al.* (1993) used fluorescence particles to trace particle movement and concluded that there is considerable lateral particle exchange between the upflow core region and the downflow wall region. Bolton & Davidson (1988), Herb *et al.* (1992) and Rhodes *et al.* (1992c) argued that there must be many particles moving from the core to the annulus. The large lateral fluxes of solids measured by Qi & Farag (1993) in the near wall region discussed above also indicate extensive radial solids mixing. These findings are also consistent with the observations by Martin *et al.* (1992) in a FCC riser reactor where a flat radial distribution profile of coke yield suggests considerable radial solids mixing.

8.5. High density flux CFBs

Most published CFB studies have been carried out at relatively low solids circulation rates ($< 200 \text{ kg/m}^2\text{s}$) and with low solids concentration in the upper region of the risers ($\overline{\epsilon} > 97\%$) corresponding to the operating conditions of circulating fluidized bed combustion (CFBC), even though many industrial circulating fluidized beds, as in fluid catalytic cracking (FCC) and other gas phase catalytic reactions, operate at much higher solids circulation rates. Zhu & Bi (1995) noted that only ten publications reported experiments with solids fluxes higher than 200 kg/m²s. Most FCC design and operation still rely mainly on in-house proprietary data and correlations (Avidan *et al.* 1990). Basic understanding of the fluidization behaviour inside FCC risers, regenerators and strippers is still lacking. Studies of high density/high flux CFBs are therefore needed.

To maintain stable CFB operation, there must be enough pressure available from the downcomer side (i.e. the pressure at the bottom of the return column minus the pressure drop across the solids feeding device) to support the solids holdup in the riser (Bi & Zhu 1993). If the pressure at the bottom of the return column is increased due to increased solids inventory or the pressure drop across the feeding device is reduced, the solids holdup on the riser side must also increase to balance the system. The pressure head of the blower must also be high enough to support the solids holdup. Slugging or classical choking of the riser must be avoided when the riser is operated under high density, high solids flux conditions (Bi *et al.* 1993b). Large riser diameters and small particles (e.g. FCC particles) are most suitable for such operation, because the maximum bubble size for these particles is generally smaller than 200 mm (Zhu & Bi 1995).

The conditions required to achieve high density are (Zhu & Bi 1995):

- (a) sufficient blower capacity and pressure,
- (b) high solids inventory,



Figure 14. Operating limits of CFB riser (Zhu & Bi 1994) for particles of diameter 60 mm and density 1500 kg/m³ for a riser of diameter 0.15 m and height 10 m. (D: riser diameter, m; D_d : diameter of solids feeder, m; D_s : diameter of standpipe, m; D_v : equivalent diameter of the mechanical solid control valve, m; L: total height of solids inventory in standpipe, m.)

- (c) appropriate geometry: large downcomer-to-riser diameter ratio, low resistance to solids recirculation and low gas-solid separator pressure drop,
- (d) small particles to avoid slugging and classical choking.

For given operating conditions, there is a maximum attainable solids circulation rate above which operation becomes unstable as shown in figure 14. Below the corresponding maximum, the solids circulation versus superficial gas velocity curve is the operating range for a given unit. The effects of standpipe-to-riser diameter ratio, static-bed-height to riser-height ratio and solids control valve setting on the operating range, calculated using the model of Bi & Zhu (1993), are also shown in figure 14. The geometry of the riser and downcomer play important roles in determining the limits of operation. The maximum solids holdup tends to increase with increasing standpipe-to-riser diameter ratio and solids inventory. A higher resistance across the solids feeding device and/or gas-solids separator can also reduce the maximum solids flux and thus the operation range.

From this analysis, Zhu & Bi (1995) explained why an FCC riser can be operated at much higher solids flux than a CFB combustor. FCC risers employ fine group A particles which are much less prone to choking than the group B solids used in CFB combustors. The FCC unit has a large diameter regenerator and thus large solids inventory to maintain catalyst recirculation, while the combustor has a much smaller standpipe and smaller solids inventory. Few laboratory-scale CFB units have been operated at high solids flux, high solids-gas feed ratio conditions, because the high solids flux can only be achieved with large solids storage beds (Yerushalmi *et al.* 1976; Galtier & Pontier 1989; Ambler *et al.* 1990; Martin *et al.* 1992), large solids inventory (Arena *et al.* 1991; Galtier & Pontier 1989), coupled with low resistance control valves or high aeration (Patience *et al.* 1991; Pugsley *et al.* 1993), conditions usually not available for laboratory-scale units.

8.6. Downer reactors

While the CFB reactor is suitable for many chemical processes, it suffers from significant solids backmixing due to solids downflow at the wall. This has led to the development of the downflow fluidized bed reactor, usually simply referred to as the 'downer reactor' in which gas and particles are made to flow concurrently downwards. This avoids all backmixing of solids and, with appropriate attention to even distribution of solids, gives very little axial dispersion of either solids or gas. An extensive review of the downer reactor is being published elsewhere (Zhu *et al.* 1995).

Uniform distribution of solids at the top of downer reactors can be achieved by having many tubes at the top of the reactor delivering particles from a fluidized bed of solids above (Gartside 1989; Qi *et al.* 1990; Aubert *et al.* 1994). The fluidized bed is kept near minimum fluidization to avoid bubbles which could block the distribution tubes. When two or more different solids are



Figure 15. Radial solids concentration in a riser of diameter 0.14 m and overall height 11 m and in a downer of diameter 0.14 m and overall height 5.8 m under similar operating conditions for different distances h from the distributor for $U_G = 2.9$ m/s and $G_s = 108$ kg/m²s (from Yang *et al.* 1991a).

required, impinging jets provide ultra-rapid mixing (Berg *et al.* 1989), although at the expense of potentially high rates of attrition.

The downer reactor consists of a cylindrical column in which the particles are initially accelerated by both drag and gravity and reach velocities which exceed the superficial gas velocity. Pressure profiles (Wang *et al.* 1992) show that the pressure initially decreases with distance from the entrance, due to the acceleration of particles caused by drag, and then increases with distance from the entrance, once the particles fall more quickly than the gas.

The radial distribution of flow in the downer is much more uniform than in the riser. Instead of the core-annulus structure described above for circulating fluidized bed risers, where there is a marked increase in solids concentration in a thin wall layer, the downer tends to show a small annular peak in solids concentration at some distance from the wall (Bai *et al.* 1991; Yang *et al.* 1991; Wang *et al.* 1992). Typical profiles are shown in figure 15 in comparison with typical riser profiles. The solids concentration is found to become more uniform as the superficial gas velocity is increased at a given solids flux and as the solids flux is decreased at a constant gas velocity.

Gas velocity profiles are nearly flat except near the wall, and there is very little variation with height (Cao *et al.* 1994). The gas velocity reaches a maximum at r/R = 0.85 to 0.96, corresponding to the position of the maximum local solids concentration. Radial distributions of particle velocity tend to be similar to the particle concentration profiles (Yang *et al.* 1991a). As a consequence, the radial distribution of solids flux is also similar, but even less uniform, being obtained from the product of local solids concentration and velocity. The downer reactor can be considered to consist of three regions: a central core of uniform particle concentration and velocity, an annular ring of increased particle concentration and augmented gas and particle velocity, and an outer wall-dominated zone of reduced velocities and particle concentration. While these variations do exist, the downer is considerably more uniform radially than corresponding risers.

Relative velocities in downers are distributed radially in a similar manner to particle concentration (Cao *et al.* 1994). These relative velocities tend to be several times the individual particle terminal velocity, suggesting some particle congregation, but less than in risers.

Axial dispersion of both gas and solids is significantly less than in comparable risers (Wei *et al.* 1994b, Zhu & Wei 1995), whereas radial gas and solids mixing appear to be of the same order.

8.7. Future research needs

While many hydrodynamic studies have been carried out and much better understanding of CFB hydrodynamics has been achieved in the last ten years, there are still many areas where further research is needed:

- (a) Most studies reported so far have employed FCC or sand particles. Systematic studies with varying particle properties are needed to broaden knowledge of CFB design.
- (b) While progress has been made in understanding the local flow structure of gas and particles (e.g. of clusters), a much better picture of the flow structure is needed to allow the development of realistic and predictive hydrodynamic models.
- (c) Transitions from turbulent to fast fluidization and from fast fluidization to pneumatic transport need to be established clearly, to delineate appropriate ranges of operation.
- (d) Systematic study is needed to elucidate the detailed flow structure of the lower dense phase region and its effect on higher regions.
- (e) As discussed, few studies have been carried out with high solids flux and high solids holdup in the riser, as in modern FCC reactors. Further research is needed.
- (f) The downer reactor shows promise for short contact time gas-solids reactions, but only very limited studies have been reported.
- (g) Most data reported in the literature are from risers smaller than 200 mm in diameter. More data are needed from large-scale units.
- (h) More accurate measurements are needed of axial and radial particle fluxes, gas velocity and average local particle velocities. The various experimental techniques should be compared to elucidate their relative advantages and disadvantages.
- (i) New robust instruments are required to measure the hydrodynamics inside industrial units operating at high temperatures and pressures.

9. AIDS TO FLUIDIZATION

For many years there have been attempts to improve the performance of fluidized beds by modifying the particle behaviour via externally applied fields (e.g. electrical, magnetic, acoustic or centrifugal) or by means of baffles. These efforts have continued in recent years, but at a diminished pace.

9.1. Magnetic fields

Although the influence of magnetic fields on fluidized beds had been studied in Eastern Europe through the 1960s and 1970s (see Penchev & Hristov 1990), interest in the West was kindled by Rosenweig (1979) who showed that the application of magnetic fields to ferromagnetic particles could greatly extend the range of bubble-free operation by raising $U_{\rm mb}$ while $U_{\rm mf}$ is unaffected.

Recent work has contributed to understanding how a co-axial magnetic field can cause group B powders to be converted to group A behaviour (Penchev & Hristov 1990), how bubble sizes can be diminished by varying the intensity of the magnetic field (Jovanovic *et al.* 1989) and how both gas and solids mixing are reduced in magnetically stabilized beds relative to normal beds (Geuzens & Thoenes 1988; Warner & Bischoff 1992).

9.2. Acoustic fields

Chirone *et al.* (1992 and 1993) have demonstrated that acoustic fields can cause disaggregation of particle clusters into smaller units composed of a few relatively large agglomerates and many smaller ones, allowing group C powders to be fluidized as if they were group A materials. In their experiments, the particle size ranged from 1 to $45 \,\mu$ m, with a Sauter mean diameter of only 11 μ m. Their work indicates that acoustical field effects extend to gas velocities well above minimum fluidization and result in much more reproducible behaviour of cohesive powders. Similar findings were reported by Nowak *et al.* (1993) who also demonstrated that acoustic energy improved the quality of fluidization once it had been achieved.

9.3. Baffles

There has been considerable work in the past showing that internal surfaces can modify fluidization behaviour. Dutta & Suciu (1992) used the largest variety of baffles ever reported (no fewer than 29 different geometries) to study their tendency to break up voids. Although the range of operating conditions covered was small, the results do indicate smaller bubbles in the presence of baffles, with the extent of break-up being a function of baffle geometry. Yates and Ruiz-Martinez (1987) employed X-ray photography to record bubbles interacting with horizontal tubes. Not only did the tubes cause bubble break-up, but the total volume of bubbles decreased as a result of the splitting process, with gas leaking into the dense phase.

10. SCALING OF FLUIDIZED BEDS

The principles of dynamic similitude can sometimes be useful in allowing experiments to be performed on a small scale in order to provide information on a full-scale system. In order to ensure that the reduced scale unit gives useful results, one requires geometric similarity and equality of all important dimensionless groups. Since an extensive review of this subject has been published recently (Glicksman *et al.* 1994), only brief comments are included here.

A number of attempts (e.g. Newby & Keairns 1986; Roy & Davidson 1989; Glicksman *et al.* 1991; Westphalen & Glicksman 1994) have been made to verify that similarity is indeed achieved in equipment of different sizes when the similarity laws are followed, using such measures as pressure fluctuations in bubbling beds or pressure profiles in circulating beds to test for dynamic similarity. These results have never provided conclusive validation, although they have generally given reasonable correspondence between corresponding systems. Full scaling requires the matching of a Reynolds number, Froude number, ρ_p/ρ_G ratio, all length scale ratios, dimensionless particle size distribution and particle shape (Glicksman 1984). At high and low Reynolds numbers, the set of groups can be reduced somewhat (Glicksman 1984 and 1988; Horio *et al.* 1986) and there has been considerable debate over alternative formulations.

While the scaling approach has some usefulness for scaling of beds of group B and D solids operated at high temperature and atmospheric pressure (as in atmospheric fluidized bed combustion), it is not helpful when scaling beds of group A solids (where interparticle forces tend to be important) or for high pressure systems (where the dynamically similar atmospheric pressure unit ends up being larger than the full-scale equipment). The approach may also miss effects like coefficients of restitution or Coulomb friction coefficient which may be important, at least for beds operating in the fast fluidization regime (Chang & Louge 1992).

11. CONCLUDING REMARKS

Recent years have seen both the extension of fluidization work from previous years and movement in new directions. Gas-solid systems continue to offer intriguing fundamental questions, difficult practical dilemmas and opportunities for innovation. The next years will continue to see evolution, particularly towards:

- (a) improving the understanding of high-velocity systems,
- (b) extending the range of particles which can be treated in fluidized systems,
- (c) novel contactors such as the downer and jet configurations,
- (d) optimizing factors like particle size distribution,
- (e) diagnostic systems able to make new or more precise measurements,
- (f) knowledge-based control systems,
- (g) advanced computer-based hydrodynamic models.

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