Effect of Particle Size Distribution in Different Fluidization Regimes

Guanglin Sun and John R. Grace
Dept. of Chemical Engineering, The University of British Columbia, Vancouver, Canada V6T 1Z4

The effect of particle size distribution (PSD) on the performance of a fluidized-bed reactor was investigated in different hydrodynamic regimes: bubbling, slugging, turbulent and fast fluidization, with three particle size distributions, all with the same mean diameter and nearly the same particle density and BET surface area. Regime transitions were examined by measuring pressure fluctuations. Void sizes tended to be smaller and the transition from bubbling or slugging to turbulent fluidization was achieved earlier for the wide distribution powder than for the narrow PSD. At gas velocities ≤ 0.2 m/s, the conversion and reactor efficiency were not affected greatly by the PSD. However, at higher gas velocities, PSD played a significant role. For particles of wide size distribution, the conversion in the turbulent and fast fluidization regimes was usually higher than in the bubbling fluidization regime at the same dimensionless rate constant, k_f'. On the other hand, for particles of narrow size distribution, the dependence of conversion on the regime is small, except for the fast fluidization regime.

Introduction

In most previous experimental studies of chemical reaction in fluidized beds, whether for group A or for group B particles (for example, Lewis et al., 1959; Hovmand et al., 1971; Chavarie and Grace, 1975; Bauer and Werther, 1981; Yates and Newton, 1986), restricted ranges of gas velocity, seldom exceeding 0.3 m/s, have been investigated. The contacting efficiency of fluidized beds tends to decrease with increasing gas velocity within this range, since the above studies have been concerned with the bubbling or slugging regime where bubbles or slugs grow larger with increasing gas flow. Most successful applications of fluidized beds in the petrochemical or chemical industries, such as fluid catalytic cracking, acrylonitrile synthesis and oxychlorination of ethylene, however, involve higher gas velocities, generally sufficiently high that clearly defined bubbling or slugging has given way to a more turbulent hydrodynamic region.

The gas velocity, U_c, at which the pressure fluctuations reach a maximum, can be taken as a "pseudo" critical gas velocity to distinguish transition to the turbulent regime of fluidization (Yerushalmi, 1986). Below U_c, the two-phase appearance of fluidized beds is clear, and voids grow larger with increasing gas flow at a rate that depends on the gas and solid properties and on the equipment characteristics. Around U_c, the heterogeneous, two-phase character of the bed reaches a maximum. Above U_c, the fluidized bed gradually gives way to a condition of increasing uniformity, culminating in a "turbulent" state. Complete transition to the turbulent regime is said to have occurred at a superficial velocity, U_k, where large discrete bubbles or voids are largely absent and the amplitude of pressure fluctuations has leveled off. The turbulent regime then extends to the transport velocity. The approach to the transport velocity is accompanied by a sharp increase in the carryover rate. The transport velocity may be regarded as the boundary between turbulent fluidization and fast fluidization if the solids flow rate is sufficiently high, or the transition to the dilute-phase flow regime if the solids flow rate is low.

Although fluidized-bed reactors have advantages for many chemical processes compared with other reactors, their application is restricted often by limited mass transfer between phases. Two general methods have been used in commercial practice to improve gas-solid contacting. One is to restrict the bubble growth and control gas flow by internal baffles or by dividing the bed into several stages in series. Another is to

Correspondence concerning this article should be addressed to J. R. Grace.

suppress the bubbling phenomena by changing such properties as mean particle size, particle size distribution (PSD), absolute pressure, and superficial gas velocity. The latter method is more attractive for most practical applications of catalytic fluidized-bed reactors.

Lanneau (1960) investigated bubbling phenomena using capacitance probes in a 75-mm-dia. fluidized bed over a wide range of gas velocity (0.03 to 1.5 m/s). He reported that the character of the signals changed with increasing gas velocity, suggesting a change in hydrodynamic regime. Kehoe and Davidson (1971) detected bubbles and slugs in 50- and 100-mm-dia. fluidized beds of fluid cracking catalyst. They described the breakdown of slugs to a so-called "turbulent" fluidization regime at gas velocities between 0.45 and 0.5 m/s. Since then, fluidization performance in different regimes, especially the transition from bubbling or slugging to turbulent fluidization, has been considered by many investigators (Massimilla, 1973; Thiel and Potter, 1977; Canada et al., 1978; Yerushalmi et al., 1978; Avidan and Yerushalmi, 1982; Grace, 1986; Jin et al., 1986; Geldart and Rhodes, 1986; Yang et al., 1988; Sun and Chen, 1989; Judd and Goosen, 1989).

Previous work has indicated that the transition velocity from bubbling to turbulent fluidization increases with increasing particle size and density (Jin et al., 1986; Sun and Chen, 1989) and decreases with increasing column size (Thiel and Potter, 1977; Sun and Chen, 1989; Judd and Goosen, 1989). Little information, however, is available on the effect of particle size distribution at high gas velocities, and results showing the effect of size distribution for solids with identical mean size are lacking (Yerushalmi, 1986). The PSD has been shown to affect the performance of fluidized-bed reactors at low gas velocities, corresponding to the bubbling and slugging fluidization regimes (Sun and Grace, 1990; Grace and Sun, 1990). It was demonstrated that this effect can be predicted using the two-phase bubbling model (Grace, 1984, 1986) modified to allow for increased concentration of particles and "fines" in the dilute phase for group A particles (Sun and Grace, 1990). In this article, we report results of the PSD effect on the fluidization regime and on the performance of catalytic fluidized-bed reactors in different hydrodynamic regimes.

Experimental Equipment and Procedure

The PSD effect on the performance of a fluidized-bed reactor in different fluidization regimes was investigated using the ozone decomposition reaction. Fluid cracking catalysts were activated by being impregnated with ferric nitrate solution and then roasted. The catalysts tested had three particle size distributions—wide, narrow and bimodal—all with very nearly the same mean diameter ($\bar{d}_p \approx 60$ μm) and nearly the same particle density and BET surface area, as shown in Figure 1 and Table 1.

The superficial gas velocity in the reactor ($H_{mf} \approx 0.7$ m) was varied from 0.1 to 1.8 m/s to include the bubbling, slugging, turbulent and fast fluidization regimes. The catalytic rate constant was measured in a fixed-bed reactor with the same range of mean gas residence time as in the fluidized bed. Pressure fluctuation measurements were carried out with two static bed heights, 0.7 and 1.0 m, and at gas velocities from 0.03 to 1.7 m/s.

The experimental fluidized-bed reactor was 0.1 m ID×2.6 m tall, while the fixed bed was 0.019 m ID×0.2 m long. Both

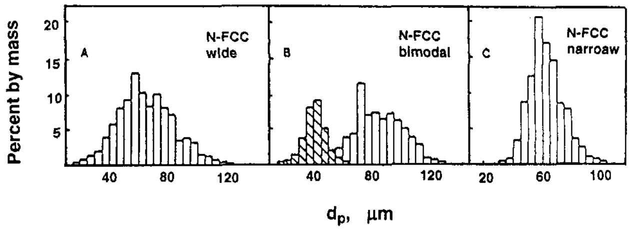

Figure 1. Size distributions of different particle blends.

reactors were constructed of aluminum to avoid catalysis by the column material. Particles collected by the cyclones (two in series) were returned to the reactor in the lower dense region, not far above the distributor, by a double-decked flapper system designed to keep the PSD within the experimental system nearly constant during experimental tests by preventing fine particles from escaping.

The void characteristics and regime transitions were determined by measuring pressure fluctuations along the column. Four differential pressure transducers were used, each with a range of 0 to 7 kPa. The tubing connecting the taps and transducers was checked experimentally to ensure that no significant damping was introduced. Differential pressure signals at different operating conditions were logged to an IBM-XT minicomputer via an analog-to-digital convertor and recorded simultaneously in analogue format on a UV chart recorder.

Differential pressure was measured along three 250-mm intervals at three heights along the column, 30 to 280 mm, 430 to 680 mm, and 680 to 930 mm. These are designated as Δz_1, Δz_2, Δz_3, respectively. For $H_{mf} = 0.7$ m, measurement taps across Δz_1 and Δz_2 were always immersed in the dense bed. For $H_{mf} = 1.0$ m, all three taps were always immersed. Pressure differences between each interval were measured for 40 s for each operating condition, with 100 data points recorded each second. Additional details are given elsewhere (Sun and Grace, 1990; Sun, 1991).

Results and Discussion

Regime transitions

The differential pressure fluctuations associated with the two-phase character of the fluidized bed allow the regime transitions from bubbling to slugging fluidization and from slugging to turbulent fluidization to be investigated. A dimensionless standard deviation defined as:

$$F_p = \frac{\sqrt{\sum_{i=1}^{N} (\Delta P_i - \overline{\Delta P})^2 / N}}{\Delta \bar{P}} \tag{1}$$

is used to express the normalized amplitude of the pressure

Table 1. Relevant Properties of Fluid Cracking Catalyst

PSD	Wide	Narrow	Bimodal
ρ_p, kg/m^3	1,580	1,570	1,585
ρ_b, kg/m^3	860	820	865
BET Area, m^2/g (Unactivated)	212	208	210
BET Area, m^2/g (Activated)	172	164	169

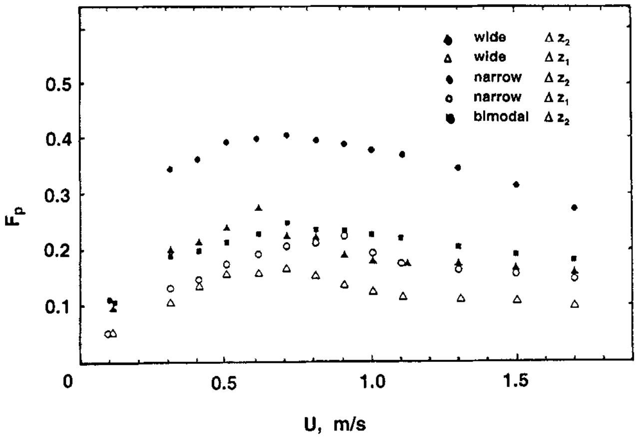

Figure 2. Effect of PSD on pressure fluctuations with $H_{mf}=0.7$ m, over intervals Δz_1 (30 to 280 mm), Δz_2 (430 to 680 mm), and Δz_3 (680 to 930 mm).

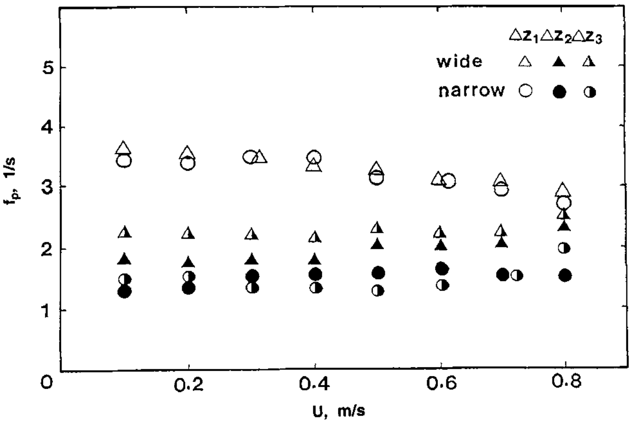

Figure 4. Influence of PSD on the frequency of pressure fluctuations in fluidized beds with $H_{mf}=0.7$ m, over intervals Δz_1, Δz_2, and Δz_3 (as in Figure 2).

drop fluctuation, where N is the total number of data points, $\Delta\bar{P}$ is the mean pressure drop across the measured section, and ΔP_i is the instantaneous pressure drop across the same section.

The variation of F_p with superficial gas velocity for three powder blends is shown in Figures 2 and 3. It is clear that the amplitude of differential pressure fluctuations was influenced by the PSD. Among the three size distributions used in our experiments, the narrow blend gives the highest F_p for given operating conditions, except at very low gas velocities ($U<0.2$ m/s), where the values of F_p were similar for the different PSDs.

A typical plot showing the influence of the PSD on the frequency of pressure fluctuations, f_p, is shown in Figure 4. To separate the noise from significant peaks in determining frequencies, peaks were counted only if their amplitude was at least 40% of the root-mean-square amplitude of pressure fluctuations, as determined by the numerator of Eq. 1. At Δz_1, the frequencies indicate the presence of the bubbling regime for $U>0.1$ m/s. In the upper part of the bed, however, lower frequencies between 1 and 2 s^{-1} suggest slug flow. This agrees with the criterion for slug flow developed by Baeyens and Geldart (1974), which suggests a minimum gas velocity for slugging of nearly 0.1 m/s for a column diameter of 0.1 m and $H_{mf}=0.7$ m.

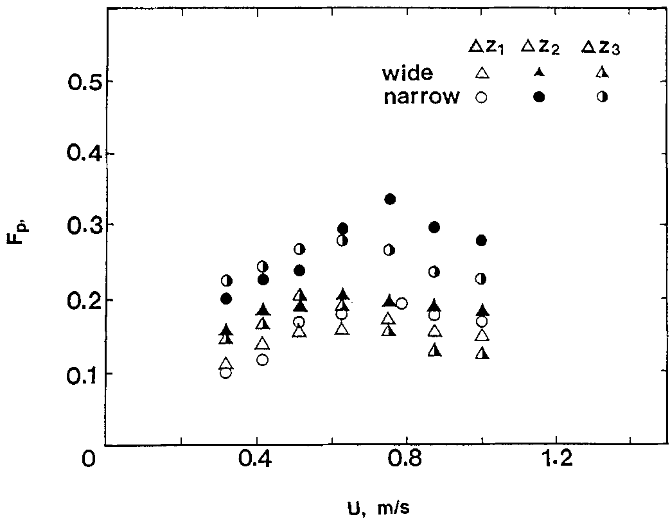

Figure 3. Effect of PSD on pressure fluctuations with $H_{mf}=1$ m, over intervals Δz_1, Δz_2, and Δz_3 (as in Figure 2).

It is notable that for all three different particle size distributions tested, the lower part of the fluidized bed ($z<0.3$ m) operated in the bubbling fluidization regime. The influence of the PSD on the bubble frequency was insignificant, consistent with the conclusion of Clift and Grace (1972). The void frequency decreased with increasing height, suggesting further bubble coalescence. Void frequency in the upper part of the bed depended on the PSD, in contrast to the lower part of the bed. In the bed of the narrow PSD, both the rate of bubble coalescence and the size of slugs, inferred from the frequency and amplitude of the pressure fluctuations, were greater than for the wide PSD.

Figures 2 and 4 show that although slugs occur in the bed for particles of three different size distributions, the slug characteristics, such as length and spacing, appear to depend on the PSD. For a fluidized bed ($H_{mf}=0.78$ m, $D_T=0.102$ m) of group A particles ($\bar{d}_p=62$ μm) with narrow size distribution, Kehoe and Davidson (1973) measured the slug spacing to be about $2D_T$. They also found that both the spacing and the slug length increased with height. These phenomena are consistent with our experimental results from the bed of the narrow PSD, where the frequency of pressure fluctuations in the upper part of the bed was usually less than 1.5 s^{-1}. In a bed of wide size distribution, voids appeared to occur as bubbles and slugs, even in the upper part of the bed. The frequency of pressure fluctuations for Δz_3 (more than 2 s^{-1}) was generally higher than that for Δz_2, suggesting some splitting of slugs or bubbles. The F_p and f_p results in Figures 2 to 4 show that higher F_p and lower f_p were achieved in the bed of the narrow PSD than for the wide and bimodal distributions. This suggests that both slug spacing and slug length are larger for a narrow PSD.

Based on the results that the average bubble size appears to be related directly to the standard deviation of the amplitude of pressure fluctuations (Kai and Furusaki, 1985; Lee and Kim,

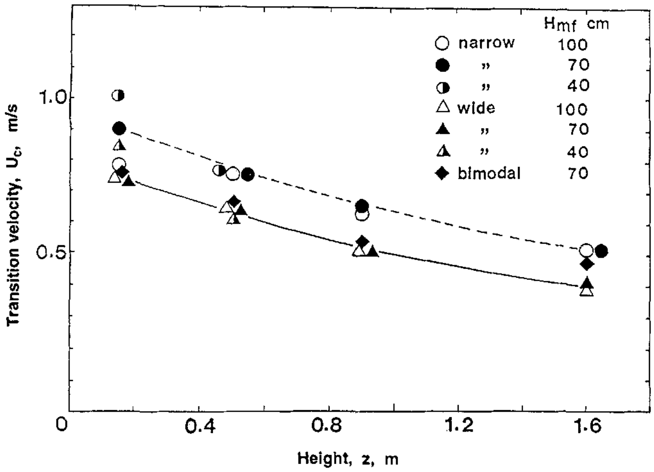

Figure 5. Effect of PSD on U_c for different H_{mf} and z.

1988), one can infer that the bubbles (or slugs) reach their maximum sizes at a superficial gas velocity of approximately U_c, where F_p reaches a maximum.

The effect of the PSD on U_c is shown in Figure 5. For the three blends of particles, the highest U_c is attained for the particles of narrow PSD, while the lowest U_c occurs for the wide distribution. The effect of static bed height on the local transition velocity was negligible, consistent with results reported by Canada et al. (1976). The local transition velocity, U_c, at which the local pressure fluctuation peaks, however, appears to decrease with height.

Fluidization performance may be influenced substantially by the effective viscosity of the dense phase (Grace, 1970). The lower the bed viscosity, the more likely that voids will split (Clift et al., 1974). One would therefore expect earlier transition from bubbling or slugging to turbulent fluidization for beds of lower viscosity. In a fluidized bed of group A particles, the effective viscosity increases with increasing mean particle diameter and decreases with increasing fines content (Kono et al., 1986). For the gas-solid system tested in our study, Khoe et al. (1991), based on collapse tests, reported that for a given average surface-volume diameter, a wide size distribution gives a greater air retention capacity than a bimodal distribution, which in turn gives greater retention than a narrow distribution.

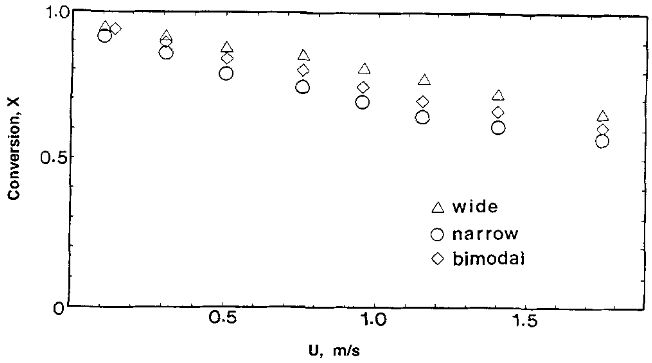

Figure 7. Influence of PSD on reactor conversions: catalyst inventory, 5 kg; $k_r \approx 9\ s^{-1}$.

The tendency we have found for the wide PSD to give smaller voids and earlier transition to turbulent fluidization than the narrow distribution is consistent with the lower effective dense-phase viscosity and greater aeration capacity that occurs as the PSD is broadened.

Reactor performance in different fluidization regimes

Experimental conversions for the three different particle size distributions are compared in Figures 6 and 7. The PSD is seen to play a more significant role at gas velocities between about 0.5 m/s and 1.4 m/s than at lower or higher gas velocities.

The influence of fluidization regime on the performance of fluidized-bed reactors was investigated by changing the gas velocity and the kinetic rate at the same time so that there were overlapping ranges of the dimensionless rate constant, k_f'. The kinetic rate constant, k_r, was varied from 2.4 to 12 s^{-1} by varying the proportions of activated and inactive FCC. For all particles under study, the effectiveness factors can be shown to be virtually unity. The conversions obtained in different fluidization regimes are shown in Figures 8 and 9 for particles of wide and narrow size distribution, respectively. The corresponding results for single-phase plug flow (PF) and for single-phase perfect mixing (PM) are also shown for reference.

Figure 8 shows that for particles of wide size distribution, the conversion obtained in the turbulent and fast fluidization

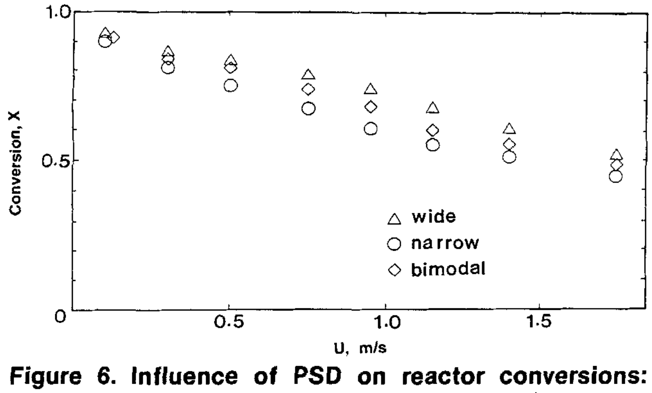

Figure 6. Influence of PSD on reactor conversions: catalyst inventory, 5 kg; $k_r \approx 4.5\ s^{-1}$.

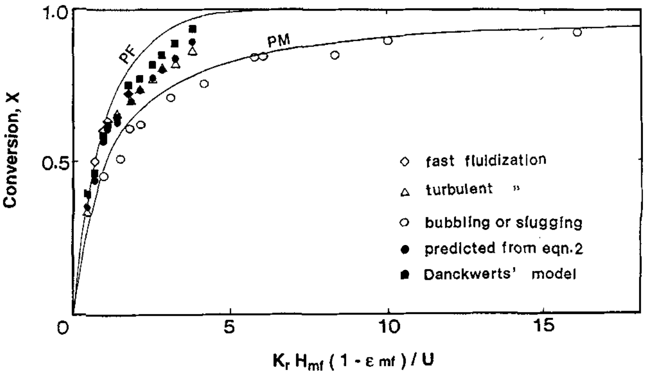

Figure 8. Influence of hydrodynamic regime on ozone decomposition for catalysts of wide size distribution.

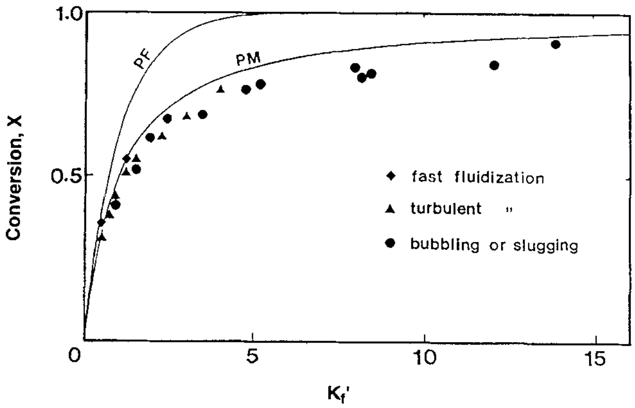

Figure 9. Influence of hydrodynamic regime on ozone decomposition for catalysts of narrow size distribution.

regimes was typically 10% to 20% more than in the bubbling and slugging fluidization regimes at the same k_f'. Conversions in the bubbling and slugging regimes were usually lower than in a single-phase, perfectly mixed reactor, suggesting significant mass transfer resistance between a dilute and a dense phase. Conversions at higher gas velocities ($U > 0.7$ m/s) in the turbulent and the fast fluidization regimes, however, were usually higher than those for corresponding perfectly mixed reactors, implying that the two-phase character or the mass-transfer resistance between phases decreases. Operating at higher gas velocities, therefore, can improve the performance of fluidized-bed reactors of group A particles of wide size distribution.

For particles of narrow size distribution, on the other hand, Figure 9 shows that there is little difference between conversions achieved at the same dimensionless kinetic rate constant, k_f', but in different hydrodynamic regimes. These trends are consistent with the measurements of pressure fluctuations described earlier and with measurements of particle concentration in voids (Sun, 1991). The influences of gas velocity and of PSD are clearly related to the two-phase characteristics of the gas-solid systems, such as the distinctiveness of two phases and mass transfer between the phases.

For particles of wide size distribution, the regime transition (U_c) from bubbling or slugging, depending on the bed height, to turbulent fluidization begins at a gas velocity of around 0.6 m/s, while the end of the transition (U_k, see Yerushalmi, 1986) is at around 1 m/s. For particles of narrow size distribution, although the pressure fluctuations at the upper part of the bed appear to decrease gradually with gas velocity after reaching a peak at around 0.75 m/s, the significant leveling off, which occurred for particles of the wide PSD, was not found for the range of conditions covered in this study. This suggests that even at high gas velocities, two-phase character is still significant for a narrow PSD, at least at the upper part of the bed. Having a proper particle size distribution is therefore essential to obtain optimum performance of fluidized-bed reactors at high gas velocities or in the "turbulent" regime.

The difference in particle concentration inside voids should also be noted. The concentration of particles inside voids is a function of a number of factors including the bubble through-flow velocity, terminal settling velocities of the individual size fractions, and disturbances at the void boundaries. For particles of wide size distribution, the solids content inside bubbles increases with increasing gas velocity for $U > 0.2$ m/s (Sun, 1991). For particles of narrow size distribution, however, solids concentration in voids usually decreases with increasing gas velocity for $U < 0.6$ m/s, in contrast to the wide PSD. More reaction therefore occurs in the voids for particles of wide size distribution than for a narrow PSD.

Modeling in different fluidization regimes

The effect of fluidization regime must be considered in modeling fluidized-bed reactors. When two-phase phenomena appear predominant, at lower gas velocities for wide PSD or over a wider range of gas velocity for narrow PSD, the conversion achieved in a fluidized bed can be successfully predicted by the two-phase bubbling bed model (Grace, 1984, 1986) with modifications to express the PSD effect on particle concentration in voids (Sun and Grace, 1990) and on the bubble and/or slug volume. At high gas velocities, however, the higher efficiency in fluidized beds of a wide PSD, with values between those obtained for single-phase, perfect-mixing and plug-flow reactors, tends to be underestimated by the two-phase bubbling bed model.

Based on experience with the MTG process, Avidan and Edwards (1986) noted that an axial dispersion model can be used to predict conversion in a turbulent fluidized-bed reactor under certain conditions: high gas velocities, sufficient fines, and sufficient bed height. These conditions were almost achieved in our experiments for particles of a wide PSD at high gas velocities. Conversions from this model were given by Danckwerts (1953) for the case of open inlet and closed outlet boundary conditions and a first-order reaction. Corresponding predictions are given in Figure 8, where the axial dispersion coefficient is based on experimental measurements by Guo (1987). Guo applied hydrogen as tracer gas to measure the residence time distribution and gas axial dispersion coefficient in a 0.07-m-dia. fluidized bed of FCC particles. The tracer was injected through a solenoid valve into the windbox of the column as an impulse input, and the sampling probe was installed in the expansion section. The measurements were carried out at superficial gas velocities from 0.1 to 1.2 m/s with H_{mf} from 0.15 to 0.9 m. These conditions are similar to those of our systems. Using the data reported by Guo for $H_{mf} = 0.6$ m, the Peclet numbers calculated for our experimental systems were from 6 to 13, similar to values measured by Avidan and colleagues (Avidan and Edwards, 1986; Krambeck et al., 1987).

Better predictions were achieved if alternate boundary conditions of closed inlet and open outlet were used, again in conjunction with axial dispersion values from Guo (1987). Details are given by Sun (1991). The expression for conversion is then given by:

$$X = 1 - \frac{2\,a\,\exp(Pe)}{(1+a)\exp[Pe(1+a)/2] - (1-a)\exp[Pe(1-a)/2]}, \tag{2}$$

where

$$Pe = HU/D_e \tag{3}$$

$$a=\sqrt{1+4k_f''/Pe} \tag{4}$$

$$k_f''=k_r H(1-\epsilon_f)/U \tag{5}$$

As shown in Figure 8, the conversions achieved in the turbulent fluidization regime for particles of wide size distribution are in good agreement with the predictions from this simple model, with an average deviation of only 5.1%, compared with an average deviation of 8.4% for the axial dispersion model with boundary conditions of open inlet and closed outlet (Danckwerts, 1953; Avidan and Edwards, 1986).

Conclusions

The transitions between fluidization regimes are influenced by the particle size distribution as well as by gas velocity and height. Both the amplitude and the frequency of pressure fluctuations are influenced by the PSD. A narrow size distribution gave the most vigorous fluctuations, while the wide blend produced the lowest. In the upper part of the bed, the lowest frequency of fluctuations occurred for the bed of a narrow PSD.

In the bubbling regime, voids appear to be smaller for a wide PSD, consistent with decreased effective dense-phase viscosity. Also, there are more particles dispersed inside the voids for a wide PSD. The influence of the PSD is the greatest at intermediate gas velocities of the order of 1 m/s corresponding to the turbulent fluidization regime. For particles of wide size distribution, the reactor performance can be improved significantly by operating in the turbulent or fast fluidization regime. However, for particles of narrow size distribution, the benefit of operating at high gas velocity is slight at best.

For beds of a wide PSD and operated at higher gas velocities, a single-phase axial dispersion model gave good predictions of the observed results,with closed and open boundary conditions at the inlet and top of the bed, respectively.

Acknowledgment

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Notation

- a = dimensionless group defined by Eq. 4
- D_e = gas axial dispersion coefficient, m^2/s
- $\bar{d}_p$ = average particle diameter, m
- D_T = bed diameter, m
- F_p = dimensionless standard deviation of the pressure drop fluctuation defined by Eq. 1
- f_p = frequency of pressure fluctuations, s^{-1}
- H = mean expanded bed height, m
- H_{mf} = bed height at minimum fluidization, m
- k_f' = dimensionless kinetic rate constant, $k_r H_{mf}(1-\epsilon_{mf})/U$
- k_f'' = dimensionless kinetic rate constant defined by Eq. 5
- k_r = first-order reaction rate constant based on the volume of the particles, s^{-1}
- N = number of data points
- Pe = Peclet number, UH/D_e
- U = superficial gas velocity, m/s
- U_c = superficial gas velocity corresponding to onset of turbulent fluidization regime, m/s
- U_k = superficial gas velocity where pressure fluctuations reach a minimum corresponding to absence or near-absence of bubbles and slugs, m/s
- X = conversion of reacting component
- z = height coordinate measured from distributor
- $\overline{\Delta P}$ = time-mean pressure drop across section of bed, N/m^2
- ΔP_i = instantaneous pressure drop across section i of bed, N/m^2
- ϵ_f = overall bed voidage
- ϵ_{mf} = bed voidage at minimum fluidization

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