# High-pressure Particulate Expansion and Minimum Bubbling of Fine Carbon Powders

Particulate expansion and minimum bubbling parameters ($n$, $u_t'$, $u_{mB}$, $\epsilon_{mB}$) are measured for fine carbon powders ($\bar{d}_p = 44$ and 112 $\mu$m) fluidized with synthesis gas ($H_2/CO = 0.8$) at pressures within the range $2{,}070 < P < 12{,}420$ kPa in an industrial, pilot-scale fluidized bed. Deviations between minimum bubbling ($u_{mB}$ and $\epsilon_{mB}$) and minimum fluidization ($u_{mf}$ and $\epsilon_{mf}$) conditions increase with increasing pressure, $P$. The expansion index, $n$, decreases with increasing $P$ and always exceeds values recommended by Richardson and Zaki for solid/liquid systems. Particulate bed expansion for the fine powders is well characterized by the equations of Foscolo et al. and Abrahamsen and Geldart. The theory of Foscolo and Gibilaro adequately estimates the onset of bubbling for both powders at all $P$, provided that experimentally determined values of $n$ and $u_t'$, are applied. For the $\bar{d}_p = 112$ $\mu$m powder, the theory of Foscolo and Gibilaro is applicable with calculated values of $u_t$ and experimental values of $n$.

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## Introduction

A number of researchers (Abrahamsen and Geldart, 1980a, b; Barreto et al., 1983; De Jong and Nomden, 1974; Geldart and Wong, 1984, 1985; Lockett and Gunnarsson, 1973; Massimilla et al., 1972; Rietema, 1973; Simone and Harriott, 1980) have found the fluidization characteristics of fine powders (i.e., Geldart, 1973, group A) at ambient conditions to deviate significantly from behavior predicted by the two-phase theory (Toomey and Johnstone, 1952). The fluidized beds expanded particulately at gas velocities exceeding the minimum fluidization velocity $u_{mf}$. The deviation of minimum bubbling velocity $u_{mB}$ and voidage $\epsilon_{mB}$ from minimum fluidization conditions increased with finer mean particle size $\bar{d}_p$ and a greater fines fraction $F$. This deviation further increased with increases in pressure $P$ (Crowther and Whitehead, 1978; Godard and Richardson, 1968; Guedes de Carvalho et al., 1978; Guedes de Carvalho, 1981; King and Harrison, 1982; Piepers et al., 1984; Rowe et al., 1982; Sciazko and Bandrowski, 1985; Sobreiro and Monteiro, 1982).

The idealized expansion of a Geldart group A powder with increasing superficial gas velocity $u_o$ is shown in Figure 1. A particulate bed expansion at gas velocities above $u_{mf}$ occurs over a limited range of $u_o$. As bubbling commences at $u_{mB}$, the overall bed voidage $\epsilon$ decreases because the volume of the dense phase in the bubbling bed is reduced more rapidly than the bubble holdup increases. This region of decreasing $\epsilon$ is an unstable bubbling region that is characterized by periodic fluctuations in bed height. A continued increase in $u_o$ results in an increasing bubble holdup and, hence, an increasing overall bed voidage $\epsilon$. This region of increasing $\epsilon$ is characterized by stable, or normal, bubbling.

The particulate bed expansion at gas velocities $u_o > u_{mf}$ can be described by the Richardson and Zaki (1954) equation:

$$u_o = u_t' \epsilon^n \tag{1}$$

where $u_t'$ is the velocity required to give $\epsilon = 1$, theoretically equal to the particle terminal velocity $u_t$. Values of the expansion index $n$ for group A gas/solid systems typically exceed those predicted by Richardson and Zaki for solid/liquid systems. Godard and Richardson (1968) calculated $n$ for the particulate expansion of 121 $\mu$m phenolic resin and 125 $\mu$m Diakon powders at pressures to $P = 1{,}414$ kPa. They found $n$ to decrease with increasing $P$, but to always exceed the predicted values. Furthermore, $u_t'$ generally exceeded $u_t$. Similar findings were made by Crowther and Whitehead (1978), who investigated the high-pressure supercritical fluidization of spherical Synclyst particles ($\bar{d}_p = 63$ $\mu$m) and angular coal particles ($19 < \bar{d}_p < 63$ $\mu$m) at pressures to $P = 6{,}900$ kPa with argon and carbon tetrafluoride

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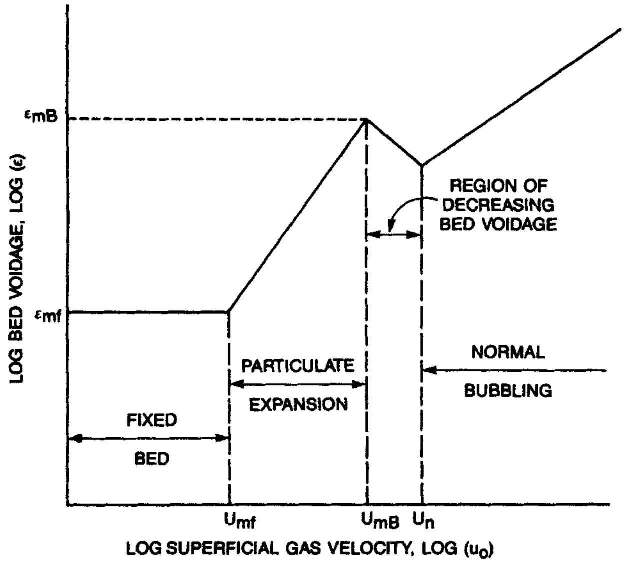


**Figure 1. Idealized bed expansion with superficial gas velocity.**
Bed of Geldart (1973) group A powder.

as the fluidizing gases. Rowe et al. (1982) evaluated $n$ and $u_t'$ for $\bar{d}_p = 70$ and 82 $\mu$m cracking catalyst powders at gas densities in the range $5.3 < \rho_g < 25.9$ kg/m$^3$. Contrary to Godard and Richardson and Crowther and Whitehead, Rowe et al. (1982) reported no effect of $P$ on $n$ or $u_t'$ and reported that calculated values of $u_t$ exceeded those of $u_t'$. Their studies were over a narrower pressure range, which may have precluded the observation of definite pressure effects.

The upper $u_o$ limit for particulate expansion corresponds to the appearance of the first bubble and is termed $u_{mB}$. Generally, $u_{mB}$ can be determined as that $u_o$ where a change in the slope of the Richardson-Zaki plot occurs, Figure 1. Although it has generally been found that $\epsilon_{mB}$ and $u_{mB}$ increase with increasing $P$ for systems of group A powders, the extent of the pressure effect and the reason for it is in question. Two theories have been proposed to explain this pressure effect phenomenon; one is based upon interparticle forces (Piepers et al., 1984), while the other is based upon hydrodynamic forces (Foscolo and Gibilaro, 1984). High-pressure ($P < 2,600$ kPa) data were analyzed by each of these groups of researchers to support their positions.

While most pressurized fluidized bed experiments to date have been limited to pressures not exceeding $P = 2,600$ kPa and to small-diameter or two-dimensional beds, the work reported here includes measurements of $n$, $u_t'$, $\epsilon_{mB}$, and $u_{mB}$ for Geldart group A carbon powders fluidized by synthesis gas in a well-instrumented, industrial, pilot-scale fluidized bed operating at pressures as high as $P = 12,420$ kPa. Parameters obtained experimentally are compared to those predicted by available correlations, and an analysis is made to determine if the Foscolo and Gibilaro theory adequately predicts the onset of bubbling.

Previous high-pressure studies with fine carbon powders fluidized by synthesis gas focused on bubbling (Weimer and Quarderer, 1984, 1985; Weimer and Jacob, 1986a) and slugging (Weimer and Jacob, 1986b) fluidized beds. The primary application of those studies and of this study is development of high-pressure Fischer-Tropsch fluidized-bed processes utilizing carbon-supported catalysts for the synthesis of liquified petroleum gases (Murchison, 1982) and mixed alcohols (*Chemical Processing*, 1985; *Chemical Week*, 1984).

## Experimental Method

### *Apparatus and instrumentation*

The experiments were carried out in a 0.097 m ID × 2.8 m, steel, fluidized bed. Feed gas is compressed and fed to the bed through a porous metal distributor. Pressure in the bed is maintained by a backpressure control valve located downstream of the bed. Gas flow rates are controlled by mass flow controllers upstream of the compressor and are verified downstream of the backpressure control valve by means of a calibrated dry test meter. The apparatus has been described in more detail previously (Weimer and Jacob, 1986a).

A nuclear density gauge (Ohmart Densart 3400; 500 mCi Cs-137 point source) is mounted to a movable scanning mechanism constructed so that the gauge can range over the entire height of the fluidized bed. The scanning mechanism is interfaced to an on-line microprocessor which controls a motor that moves the density gauge to specified locations. The on-line microprocessor monitors and performs calculations on the bed density signal. The application of this type of gauge is described in detail elsewhere (Weimer et al., 1985).

Differential pressure measurements are available at various intermediate bed heights.

### *Physical properties of powder and gas*

The fluidized powders are ground and screened activated carbon powders having mean particle sizes of $\bar{d}_p = 44$ and 112 $\mu$m. Both powders are of a fairly wide size distribution, with the $\bar{d}_p = 44$ $\mu$m powder having a fraction of fines <45 $\mu$m of $F = 0.295$.

A particle density of $\rho_p = 850$ kg/m$^3$ is carefully determined by Hg displacement with larger size fractions of the same powder ($\bar{d}_p > 500$ $\mu$m). The difficulty of accurately measuring the density of finely divided ($\bar{d}_p < 100$ $\mu$m) porous particles directly by pycnometry has been described in detail (Knight and Rowe, 1980; Knight et al., 1980) and is not attempted here. Rather, particle density, $\rho_p$, is determined by Hg displacement at atmospheric pressure for three size fractions of the activated carbon: $\bar{d}_p = 3,380$ $\mu$m (−4, +10 mesh), $\bar{d}_p = 2,275$ $\mu$m (−6, +16 mesh), and $\bar{d}_p = 1,138$ $\mu$m (−12, +30 mesh). In all cases, $\rho_p = 850$ kg/m$^3$.

Examination of the activated carbon pore size distribution, as obtained from a water desorption isotherm (Juhola and Wiig, 1949), indicates that a large portion of the micropore volume is in pores of 15 to 20 Å (1.5–2.0 nm) diameter. In addition to the microscope structure, the activated carbon is permeated by a system of macropores having a mean diameter of 800 Å (0.08 $\mu$m). No macropores larger than 2,000 Å (0.2 $\mu$m) are detected by transmission electron microscopy, thus ensuring that Hg does not enter the macropores.

Mean particle sphericity is determined by image analysis utilizing a Kontron Corp. SEM-IPS (scanning electron microscope image processing system) to be $\phi = 0.73$.

The fluidizing medium is a synthesis gas mixture ($H_2/CO = 0.8$) having an average molecular weight of $M_w = 17$ and a viscosity of $\mu_g = 1.66 \times 10^{-5}$ Pa · s at $T = 293$ K. All experiments are carried out at ambient temperature. Particle and gas physical properties are summarized in Table 1.

**Table 1. Physical Properties of Fluidization System**

| Gas Properties | |
|---|---|
| Type: Synthesis gas ($H_2/CO$ = 0.8) | |
| Avg. mol. wt., $M_w$ = 17 | |
| Viscosity, $\mu_g$ = $1.66 \times 10^{-5}$ Pa · s | |
| Temperature, $T$ = 293 K | |
| Density, $\rho_g$ kg/m³ | at Press., $P$ kPa |
| 14.20 | 2,070 |
| 28.41 | 4,140 |
| 42.61 | 6,210 |
| 56.81 | 8,280 |
| 71.01 | 10,350 |
| 85.22 | 12,420 |

Particle Properties

Type: Granular carbon
Density, $\rho_p$ = 850 kg/m³
Sphericity, $\phi$ = 0.73

Size Distribution

| Wt. % | Through µm | On µm | Wt. % | Through µm | On µm |
|---|---|---|---|---|---|
| 0.50 | 105 | 88 | 0.20 | 177 | 150 |
| 0.48 | 88 | 74 | 46.60 | 150 | 125 |
| 15.15 | 74 | 63 | 35.28 | 125 | 105 |
| 30.87 | 63 | 53 | 5.20 | 105 | 88 |
| 23.46 | 53 | 44 | 2.55 | 88 | 74 |
| 17.92 | 44 | 38 | 7.41 | 74 | 63 |
| 11.62 | 38 | 0 | 2.13 | 63 | 53 |
| 100.00 | — | — | 0.49 | 53 | 44 |
| | | | 0.12 | 44 | 38 |
| | | | 0.05 | 38 | 0 |
| $\bar{d}_p$ = 44 µm | | | 100.00 | — | — |
| | | | | $\bar{d}_p$ = 112 µm | |

## Measurements and Analysis of Results

The principle bed parameter determined throughout this study is the expanded bed height $L_f$, which is consistently determined at all $P$ and $u_o$ through application of the nuclear density gauge (Weimer et al., 1985). Increasing $L_f$ with increasing $u_o$ results from increasing $\epsilon$ brought about by an increasing dilute phase $\delta_B$ and/or increasing dense phase $\delta_D$ volume fraction. In any event, the measured bed density $\langle \rho_{bed} \rangle_c$ and differential pressure $\Delta P$ at localized points within the bed decrease as the suspended mass of solid per unit bed volume decreases ($L_f$ increases). The relationship between $\langle \rho_{bed} \rangle_c$, $\Delta P$, $L_f$, and $u_o$ is exemplified in Figure 2, where data are presented at $P$ = 8,280 kPa for the $\bar{d}_p$ = 112 µm powder. The determination of $L_f$ is generally carried out at all $P$ for $u_{mf} < u_o < u_n$, i.e., the superficial gas velocity at which normal bubbling commences. For all measurements, $L_f$ is determined as $u_o$ decreases from higher values.

### *Region of particulate expansion*

The expanded bed height, $L_f$, is determined as it varies with $u_o$ over the range of particulate expansion for both powders. With the total inventory of powder, $W_T$, in the bed known, $\epsilon$ is determined from $L_f$ as

$$\epsilon = 1 - \frac{W_T}{\rho_p L_f A_T} \tag{2}$$

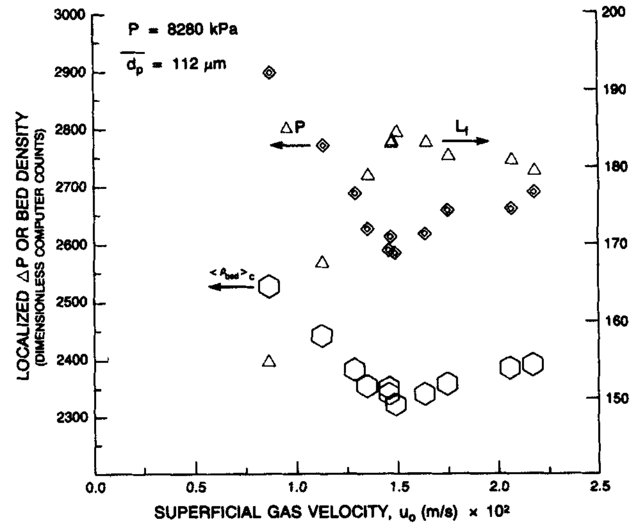


**Figure 2. Relationship of gas velocity, pressure drop, localized density, and bed height for $u_{mf} < u_o < u_n$.**

Generally, $\epsilon$ increases with increasing $P$ at equivalent $u_o$ and the rate at which the particulately fluidized bed expands with increasing $u_o$ increases with increasing $P$ for both powders, Figure 3.

Superficial gas velocity $u_o$ and $\epsilon$ are correlated closely with a Richardson and Zaki type equation, Eq. 1, for each powder at each $P$, Figure 4. Values of $n$ and $u_t'$ for each case are given in Table 2. The expansion index $n$ decreases with increasing $P$ for both powders and exceeds the value calculated from the Richardson and Zaki formulas for solid/liquid systems. The effect of $P$ is greater for the $\bar{d}_p$ = 44 µm powder as $n$ decreases 26%, from $n$ = 7.05 at $P$ = 2,070 kPa to $n$ = 5.20 at $P$ = 12,420 kPa. Over this same pressure range, $n$ decreases only 7.5% (from $n$ = 4.38 at $P$ = 2,070 kPa to $n$ = 4.05 at $P$ = 12,420 kPa) for the $\bar{d}_p$ = 112 µm powder. The extrapolated values of $u_t'$ (i.e., at $\epsilon$ = 1) for all $P$

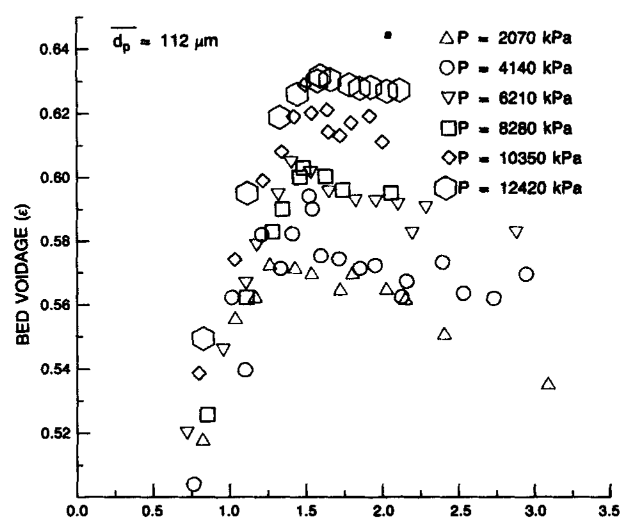


**Figure 3a. Effect of pressure and gas velocity on bed voidage for $u_{mf} < u_o < u_n$; $\bar{d}_p$ = 112 µm.**

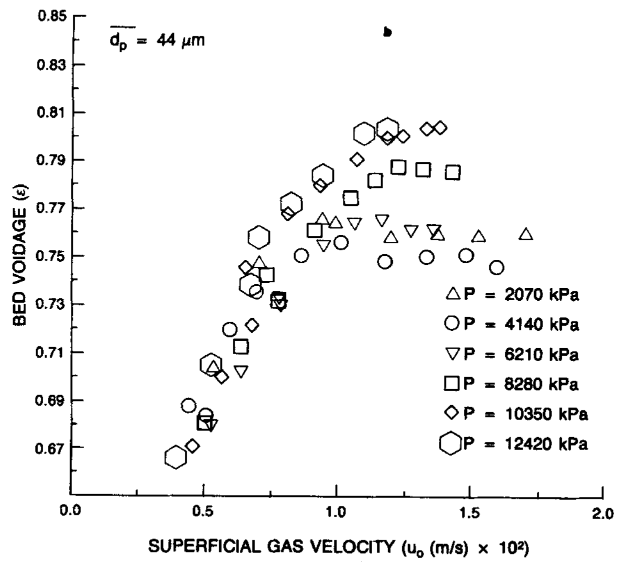


**Figure 3b. Effect of pressure and gas velocity on bed voidage for $u_{mf} < u_o < u_n$; $\bar{d}_p = 44\ \mu m$.**

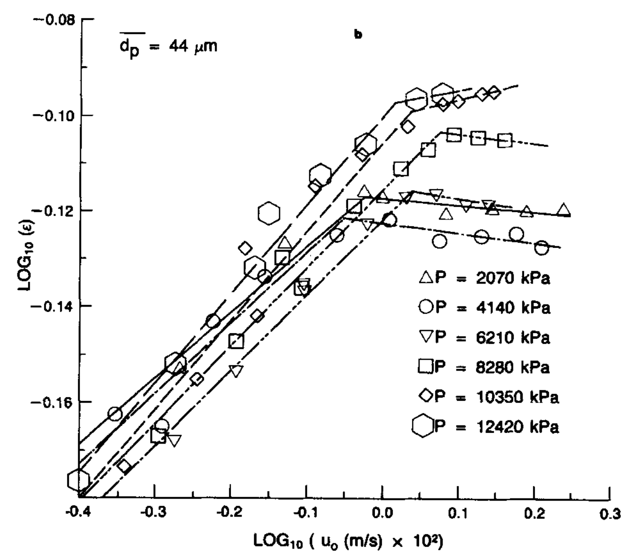


**Figure 4b. Log plot of gas velocity vs. bed voidage for data of Figure 3b; $\bar{d}_p = 44\ \mu m$.**

exceed calculated values, Table 4, for the 44 μm powder, but approximate closely those calculated for the 112 μm powder. Overall, $n$ and $u_t'$ are fitted to within ±13% for both powders at all $P$ by:

$$n = 8.115 Re_t'^{-0.1884} \tag{3}$$

## *Region of decreasing bed voidage*

The upper $u_o$ limit for particulate expansion corresponds to the appearance of the first bubble, i.e., $u_{mB}$. For increasing $u_o$ above $u_{mB}$, the bed voidage $\epsilon$ ideally decreases with increasing $u_o$, Figure 1, because the voidage of the dense phase $\epsilon_D$ in the bubbling bed is reduced more rapidly than the bubble holdup $\delta_B$ increases. For the two powders investigated here, the reduction in bed voidage is more extensive at all $P$ for the 112 μm powder, Figures 3a and 4a, than for the 44 μm powder, Figures 3b and 4b. At $P$ = 4,140 kPa, bed voidage decreases 5.4% for the 112 μm powder, from $\epsilon = 0.595$ at $u_o = 0.0154$ m/s to $\epsilon = 0.563$ at $u_o = 0.0274$ m/s. For this same pressure, bed voidage decreases only 1.3% for the 44 μm powder, from $\epsilon = 0.757$ at $u_o = 0.01009$ m/s to $\epsilon = 0.747$ at $u_o = 0.0159$ m/s. This results from the formation of larger and faster rising bubbles in the 112 μm powder, relative to the 44 μm powder, and the accompanying dominant effect of reduced $\epsilon_D$ for the 112 μm powder system.

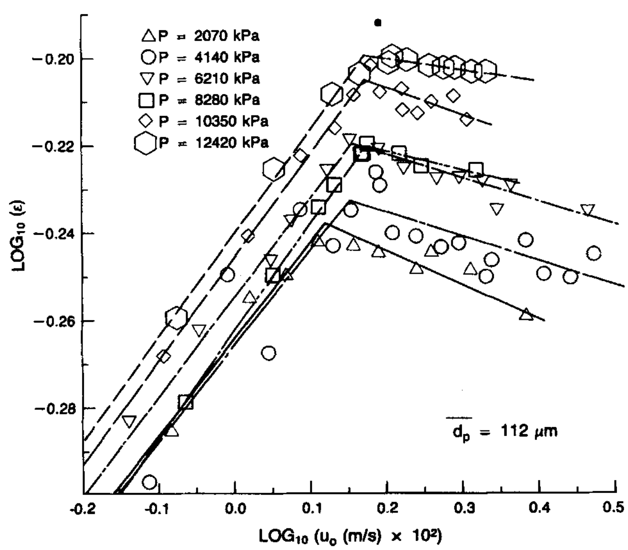


**Figure 4a. Log plot of gas velocity vs. bed voidage for data of Figure 3a; $\bar{d}_p = 112\ \mu m$.**

The effect of increasing $P$ for both powders is to reduce the magnitude of the bed voidage reduction and reduce the range of $u_o$ over which the reduction occurs. For the $\bar{d}_p = 112\ \mu m$ powder, the 5.4% voidage reduction at $P$ = 4,140 kPa is diminished to a 1.3% reduction at $P$ = 8,280 kPa (from $\epsilon = 0.604$ to $\epsilon = 0.596$) and a 0.8% reduction at $P$ = 12,420 kPa (from $\epsilon = 0.633$ to $\epsilon = 0.628$). Likewise, the range of $u_o$ over which this reduction takes place decreases as well.

In the $\bar{d}_p$ = 44 μm powder case, there is some $P$, within the range 8,280 < $P$ < 10,350 kPa, where bed voidage, rather than

**Table 2. Experimental Bed Expansion Measurements**

| Press., $P$ kPa | Index, $n$ | $u_t'$ at $\epsilon = 1$ m/s × 10² | $\epsilon_{mB}$ | $u_{mB}$ m/s × 10² |
|---|---|---|---|---|
| | | $\bar{d}_p$ = 44 μm | | |
| 2,070 | 7.05 | 6.18 | 0.765 | 0.935 |
| 4,140 | 6.58 | 5.52 | 0.757 | 0.882 |
| 6,210 | 6.16 | 5.52 | 0.767 | 1.074 |
| 8,280 | 5.95 | 4.78 | 0.790 | 1.170 |
| 10,350 | 5.22 | 3.49 | 0.798 | 1.073 |
| 12,420 | 5.20 | 3.24 | 0.801 | 1.024 |
| | | $\bar{d}_p$ = 112 μm | | |
| 2,070 | 4.38 | 14.40 | 0.578 | 1.306 |
| 4,140 | 4.36 | 14.53 | 0.586 | 1.405 |
| 6,210 | 4.27 | 12.14 | 0.604 | 1.414 |
| 8,280 | 3.90 | 10.55 | 0.603 | 1.471 |
| 10,350 | 4.02 | 9.63 | 0.625 | 1.460 |
| 12,420 | 4.05 | 9.35 | 0.633 | 1.468 |

Table 3. Estimated Minimum Fluidization Properties

| Press., $P$ kPa | $u_{mf}$, est.* m/s × $10^2$ | $\epsilon_{mf}$, calc.** |
|---|---|---|
| | $\bar{d}_p = 44\ \mu m$ | |
| 2,070 | 0.110 | 0.565 |
| 4,140 | 0.106 | 0.548 |
| 6,210 | 0.103 | 0.524 |
| 8,280 | 0.101 | 0.523 |
| 10,350 | 0.099 | 0.505 |
| 12,420 | 0.097 | 0.510 |
| | $\bar{d}_p = 112\ \mu m$ | |
| 2,070 | 0.690 | 0.500 |
| 4,140 | 0.572 | 0.476 |
| 6,210 | 0.539 | 0.482 |
| 8,280 | 0.530 | 0.464 |
| 10,350 | 0.521 | 0.484 |
| 12,420 | 0.512 | 0.488 |

*Estimated from previous experimental results (Weimer and Quarderer, 1985) and Ergun (1952) equation
**Calculated from Eq. 1 where $u_o = u_{mf}$ and with $n$, $u_t'$ from Table 2

decrease, actually increases immediately for $u_o > u_{mB}$, Figures 3b and 4b. It appears from this observation that the first bubble is not accompanied by a decrease in $\epsilon$ for operation above this pressure for the 44 $\mu m$ powder. One may speculate that this results from the formation of extremely small bubbles (1–3 mm size), as was observed by Crowther and Whitehead (1978) for very high gas density fluidization of coal particles. These investigators also reported that dense phase voidage in the bubbling bed appeared to be close to that at minimum bubbling, i.e., $\epsilon_{mB}$. This large and nearly unaffected dense phase voidage, along with the small rising velocity of such small bubbles, will result in an unaffected dense phase holdup combined with an increasing bubble holdup, and hence, an expanding bed, i.e., increasing $\epsilon$.

### *Minimum bubbling velocity and voidage*

The minimum bubbling velocity, $u_{mB}$, and, voidage $\epsilon_{mB}$, are taken as those at the point of intersection shown in Figures 4a and 4b (intersection of best-fit straight lines through bed expansion data for the regions of particulate expansion and decreasing bed voidage plotted according to Eq. 1. The experimentally determined values are summarized in Table 2 where the general trend is for $u_{mB}$ and $\epsilon_{mB}$ to increase with increasing $P$ for both powders. For the 112 $\mu m$ powder, $\epsilon_{mB}$ increases 10% over the $P$ range, from $\epsilon_{mB} = 0.578$ at $P = 2{,}070$ kPa to $\epsilon_{mB} = 0.633$ at $P = 12{,}420$ kPa. The corresponding increase in $u_{mB}$ with $P$ is from $u_{mB} = 0.0131$ m/s at $P = 2{,}070$ kPa to $u_{mB} = 0.0147$ m/s at $P = 12{,}420$ kPa. Likewise, for the $\bar{d}_p = 44\ \mu m$ powder, $\epsilon_{mB}$ increases 5% from $\epsilon_{mB} = 0.765$ at $P = 2{,}070$ kPa to $\epsilon_{mB} = 0.801$ at $P = 12{,}420$ kPa.

The effect of $P$ on $u_{mB}$ for the 44 $\mu m$ powder is somewhat different than for the 112 $\mu m$ powder. The minimum bubbling velocity $u_{mB}$ increases slightly within the pressure range $2{,}070 < P < 8{,}280$ kPa, but then appears to start decreasing with higher pressures at some pressure $8{,}280 < P < 10{,}350$ kPa. This phenomenon is concurrent with the previously reported increase in $\epsilon$ that occurs under the same conditions (i.e., appearance of first bubble not resulting in bed height reduction). However, these results differ from those of Crowther and Whitehead in that they never reported a decreasing $u_{mB}$ with increasing $P$. One should note that observations similar to those being reported here were reported, although not discussed, by Chitester et al. (1984) for $\bar{d}_p = 102\ \mu m$ ballotini and $\bar{d}_p = 157\ \mu m$ char powders fluidized with nitrogen at pressures to $P = 6{,}306$ kPa. Chitester et al. reported from visual observation of the bed that a turbulent flow regime was reached at lower gas velocities as the pressure was increased. Nonetheless, they reported no connection between this observation and the fact that the apparent $u_{mB}$ decreased with $P$. This phenomenon needs to be investigated more fully in future high-pressure work.

The results being reported here for the $\bar{d}_p = 112\ \mu m$ carbon powder are consistent with previous results (Weimer and Quarderer, 1985) for a slightly different size ($\bar{d}_p = 108\ \mu m$) carbon powder fluidized under similar conditions. In that work, collapsed bed experiments were reported for pressures to $P = 8{,}300$ kPa: $\epsilon_D$ and $u_{Do}$ were in the approximate ranges $0.50 < \epsilon_D < 0.52$ and $0.011 < u_{Do} < 0.012$ m/s, for $2{,}100 < P < 8{,}300$ kPa. These approximate ranges for $\epsilon_D$ and $u_{Do}$ are between the approximate minimum bubbling ranges ($0.58 < \epsilon_{mB} < 0.60$, $0.013 < u_{mB} < 0.015$ m/s) reported here and the minimum fluidization values where $u_{mf}$ and $\epsilon_{mf}$ decrease slightly with increasing $P$($0.50 > \epsilon_{mf} > 0.46$, $0.007 > u_{mf} > 0.005$ m/s). A similar quantitative comparison of the $\bar{d}_p = 44\ \mu m$ ($F = 0.295$) powder with the previous (Weimer and Quarderer, 1985) $\bar{d}_p = 66\ \mu m$ ($F = 0$) powder cannot be made due to significant differences in $F$ for these powders.

## Discussion

### *Estimating particulate bed expansion*

Expansion of particulately fluidized spherical solids in liquids is well characterized by the Richardson and Zaki equation (i.e., Eq. 1) where $n$ is a function of the terminal Reynolds' number $Re_t$ and the ratio $d_p/d_T$:

$$n = [4.4 + 18(d_p/d_T)]Re_t^{-0.1} \tag{4}$$

for the case of $1 < Re_t < 200$. Although it has generally been concluded that $n$ has a higher value for gas/solid systems than that of Eq. 4 (Richardson, 1971), Eq. 4 in conjunction with Eq. 1 and experimental values for $u_t'$ (i.e., $Re_t = Re_t'$) may be used to yield an initial approximation of bed expansion. The calculated relationship between $u_o$ and $\epsilon$ from Eqs. 1 and 4, using the experimentally extrapolated values for $u_t'$, Table 2, is shown in Figures 5 and 6 for the $\bar{d}_p = 112\ \mu m$ and $\bar{d}_p = 44\ \mu m$ powders, respectively, at $P = 4{,}140$, 8,280, and 12,420 kPa. Clearly, $\epsilon$ is underestimated to a large degree due to the inaccuracy of Eq. 4 for estimating $n$ for the high-pressure gas/solid particulate systems being reported here. These results are consistent with those of previous investigations reported in the literature.

Foscolo et al. (1983) derived a unified model (all voidages and flow regimes) for particulate expansion in fluidized beds which for the present intermediate flow conditions ($0.2 < Re_t < 500$) is:

$$\frac{u_o}{u_t} = \frac{[0.0777Re_t(1 + 0.0194Re_t)\epsilon^{4.8} + 1]^{0.5} - 1}{0.0388Re_t} \tag{5}$$

Again, using experimentally extrapolated values for $u_t'$ (i.e., $u_t = u_t'$ as given in Table 2), the velocity vs. voidage relationship for particulate expansion according to Eq. 5 is presented in Figures 5 and 6 and compared with the experimental relationship. The relationship estimated for the 112 $\mu m$ powder, Figure 5, is rea-

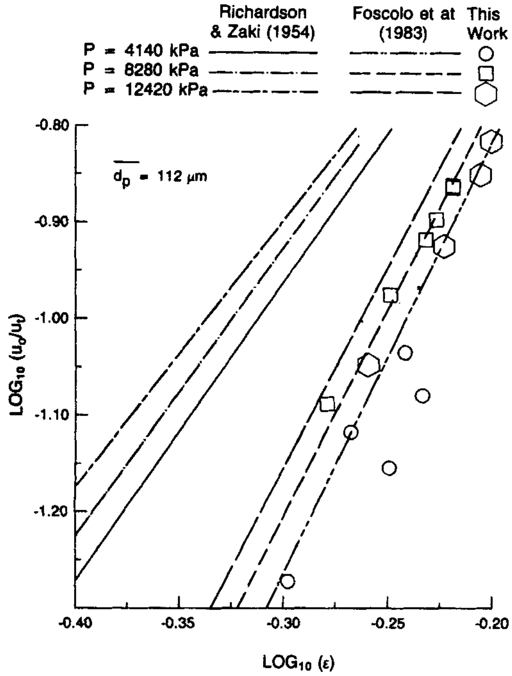


**Figure 5. Particulate expansion velocity vs. voidage relationships.**

Eq. 1 (R&Z), Eq. 5 (F e.a.), and exp. data
$u_t = u_t'$; calculated $n$; $\bar{d}_p = 112\ \mu m$

sonably accurate for all $P$, clearly superior to the Richardson and Zaki estimate. Equation 5 does not account for the significant effect fines ($F$) are known to have on expansion, and this is most likely the reason for a less accurate estimate of expansion for the 44 $\mu$m powder ($F = 0.295$) at all $P$, Figure 6.

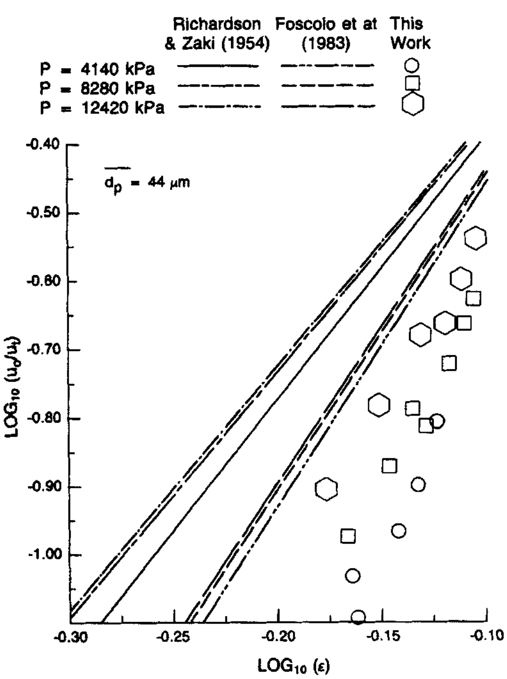


**Figure 6. Particulate expansion velocity vs. voidage relationships.**

Eq. 1 (R&Z), Eq. 5 (F e.a.), and exp. data
$u_t = u_t'$; calculated $n$; $\bar{d}_p = 44\ \mu m$

Abrahamsen and Geldart (1980a) derived an empirical expression for particulate expansion vs. velocity that accounts for the large effect of $F$:

$$\frac{\epsilon^3}{(1-\epsilon)}\frac{(\rho_p-\rho_g)g\bar{d}_p^2}{\mu_g} = 210\,(u_o - u_{mf}) + \frac{\epsilon_{mf}^3}{(1-\epsilon_{mf})}\frac{(\rho_p-\rho_g)g\bar{d}_p^2}{\mu_g} \tag{6}$$

Since Eq. 6 is correlated from experiments with group A powders having $\bar{d}_p < 75\ \mu m$, it is applied here for estimating the expansion characteristics of the $\bar{d}_p = 44\ \mu m$ powder only. It is apparent from Figure 7 that Eq. 6 provides a reasonable approximation of the high-pressure expansion reported here. Estimated $\epsilon$ is superior to that estimated using other equations, particularly at the highest pressure, $P = 12{,}420$ kPa. The disadvantage of Eq. 6 is that it requires estimated or experimental values for minimum fluidization conditions at pressure. For this study, $u_{mf}$ is approximated by adjusting experimentally determined values (Weimer and Quarderer, 1985) for similar carbon powders at high, but somewhat lower $P$. Then, values of $\epsilon_{mf}$ are estimated by using the approximations for $u_{mf}$ and Eq. 1, along with the experimentally determined values for $n$ and $u_t'$ given in Table 2. The resulting estimated values of $\epsilon_{mf}$ and $u_{mf}$ are listed in Table 3.

## *Estimating minimum bubbling velocity*

Abrahamsen and Geldart (1980b) observed that the minimum bubbling velocity, $u_{mB}$, and hence the region of particulate expansion was increased by adding fines (increased $F$) to a fluidized bed of group A powder. They proposed that:

$$u_{mB} = \frac{2{,}300\rho_g^{0.126}\mu_g^{0.523}\exp(0.716F)u_{mf}}{\bar{d}_p^{0.8}g^{0.934}(\rho_p-\rho_g)^{0.934}} \tag{7}$$

$u_{mB}$ for the 44 $\mu$m powder at all $P$ is estimated from Eq. 7, where $u_{mf}$ is given in Table 3. It is clear from Figure 8 that experimen-

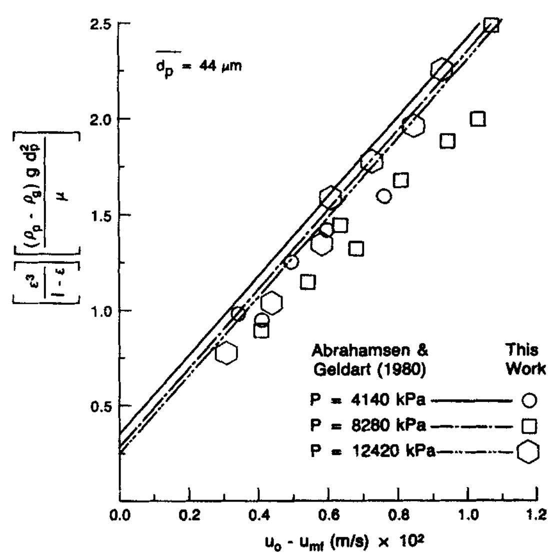


**Figure 7. Particulate expansion velocity vs. voidage relationship.**

Eq. 6 (A&G) and exp. data; $\bar{d}_p = 44\ \mu m$

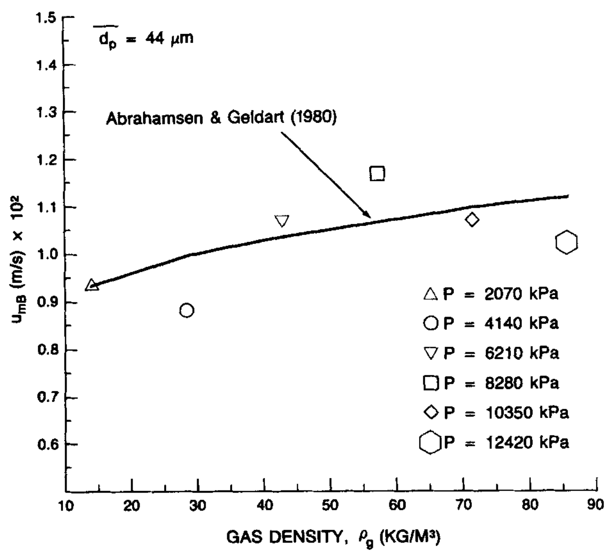


**Figure 8. Effect of gas density on minimum bubbling velocity.**
Eq. 7 (A&G) and exp. data; $\bar{d}_p = 44\ \mu m$

tal and estimated values are reasonably close, thus confirming the applicability of Eq. 7 here. For all $P$, estimated $u_{mB}$ is within ±13% of all experimental values and within ±3% of the experimental values for the $P = 2{,}070$, 6,210, and 10,350 kPa cases.

### *Estimating minimum bubbling voidage*

One theory to explain the phenomenon of particulate expansion and the effects of pressure on expansion is based upon hydrodynamic forces. Gibilaro et al. (1985) developed a general expression for the hydrodynamic interactions between particles and fluids. This applies to fixed and fluidized beds of particles for all voidages and flow regimes and led to the development of a verified model, Eq. 5, for particulate expansion in fluidized beds (Foscolo et al., 1983). These authors (Foscolo and Gibilaro, 1984) developed a general expression that led to a fully predictive criterion for the transition to bubbling. This criterion (Wallis, 1969) proposes that the stability of two-phase flow is governed entirely by the relative magnitude of the voidage propagation velocity (the speed with which a voidage change can travel through the system) and the elastic wave velocity (the speed with which particles interact together). Hence, bubbling in a fluidized bed commences when the voidage propagation velocity exceeds the elastic wave velocity. According to Foscolo and Gibilaro (1984), bubbles will first appear in a fluidized bed when:

$$\left[\left(\frac{g\bar{d}_p}{u_t^2}\right)\left(\frac{\rho_p - \rho_g}{\rho_p}\right)\right]^{0.5} = 0.56n(1 - \epsilon_{mB})^{0.5}\epsilon_{mB}^{n-1} \qquad (8)$$

This is a remarkably simple relationship for predicting the minimum bubbling voidage, $\epsilon_{mB}$, and requires only the physical properties of the system, the single particle terminal velocity $u_t$, and the expansion index $n$. Equation 8 was successfully applied (Foscolo and Gibilaro, 1984) to predict $\epsilon_{mB}$ for various pressurized gas fluidized systems, however, these were limited to pressures less than $P = 2{,}600$ kPa. In view of the wide applicability of Eq. 8, a primary objective of this work is to thoroughly evaluate its validity for the high-pressure (to $P = 12{,}420$ kPa) systems studied here.

The validity of Eq. 8 is evaluated for:

1. The case of experimentally determined $n$ and $u_t'$, Table 2; i.e. $u_t = u_t'$
2. The case of *a priori* application where $u_t$ and $n$ are calculated
3. The case where $u_t$ are calculated and $n$ are determined experimentally.

In the first case (experimental $n$, $u_t'$), correspondence between experimental values of $\epsilon_{mB}$ and those estimated by Eq. 8 are quite good. For the 44 μm powder, Table 4 and Figure 9a, experimental values for $\epsilon_{mB}$ are slightly greater (within 4%) than the estimated values for pressures at or below $P = 8{,}280$ kPa. For higher pressures, the situation reverses and estimated $\epsilon_{mB}$ exceed the experimental values. For the 112 μm powder, Table 1 and Figure 9b, experimental values of $\epsilon_{mB}$ are slightly greater than the estimated values at all $P$. Correspondence improves with increasing $P$. For all $P$ with both powders the average deviation between experimental and estimated values is 3%. This is quite a good correspondence in view of the experimental difficulty in accurately and consistently measuring $\epsilon_{mB}$.

In the second case (calculated $n$, $u_t$), the correspondence between experimental values, Table 2, and estimated values, Table 4, is poor. For the 112 μm powder at $P = 2{,}070$ kPa, the experimental value ($\epsilon_{mB} = 0.578$) exceeds the calculated value ($\epsilon_{mB} = 0.470$) by 23%. Correspondence improves with increasing $P$. For the 44 μm powder at all $P > 2{,}070$ kPa, Eq. 8 incorrectly estimates fluidization to be particulate at all $u_o$.

In the third case (experimental $n$, calculated $u_t$), correspondence between experimental values, Table 2, and calculated values, Table 4, of $\epsilon_{mB}$ for the 112 μm powder is good at all $P$. This results from the apparent validity of Eq. 8 along with good correspondence between experimental $u_t'$ and calculated $u_t$ for this

**Table 4. Calculations for Applying Foscolo-Gibilaro (1984) Theory to Predict $\epsilon_{mB}$**

| Press., $P$ kPa | Index, $n$ calc. | $u_t$,** calc. m/s × 10² | $\epsilon_{mB}$ Calc. by Eq. 8: Case 1 Exp. $n$*** Exp. $u_t'$*** | Case 2 Calc. $n$* Calc. $u_t$** | Case 3 Exp. $n$*** Calc. $u_t$** |
|---|---|---|---|---|---|
| | | $\bar{d}_p = 44\ \mu m$ | | | |
| 2,070 | 4.24 | 3.93 | 0.744 | 0.812 | 0.829 |
| 4,140 | 4.00 | 3.50 | 0.750 | partic. | 0.854 |
| 6,210 | 3.87 | 3.25 | 0.737 | partic. | 0.886 |
| 8,280 | 3.79 | 3.04 | 0.760 | partic. | partic. |
| 10,350 | 3.72 | 2.87 | 0.849 | partic. | partic. |
| 12,420 | 3.68 | 2.71 | partic. | partic. | partic. |
| | | $\bar{d}_p = 112\ \mu m$ | | | |
| 2,070 | 3.40 | 14.55 | 0.559 | 0.470 | 0.557 |
| 4,140 | 3.23 | 12.16 | 0.554 | 0.495 | 0.592 |
| 6,210 | 3.14 | 10.74 | 0.584 | 0.520 | 0.613 |
| 8,280 | 3.08 | 9.75 | 0.591 | 0.544 | 0.613 |
| 10,350 | 3.03 | 8.99 | 0.622 | 0.567 | 0.642 |
| 12,420 | 3.00 | 8.38 | 0.629 | 0.591 | 0.663 |

Partic.: particulate
*From Eq. 4, utilizing calculated $u_t$**
**Calculated from Clift, et al. (1978) for $\phi = 1$, corrected for $\phi = 0.73$ from Geldart (1986)
***From Table 2

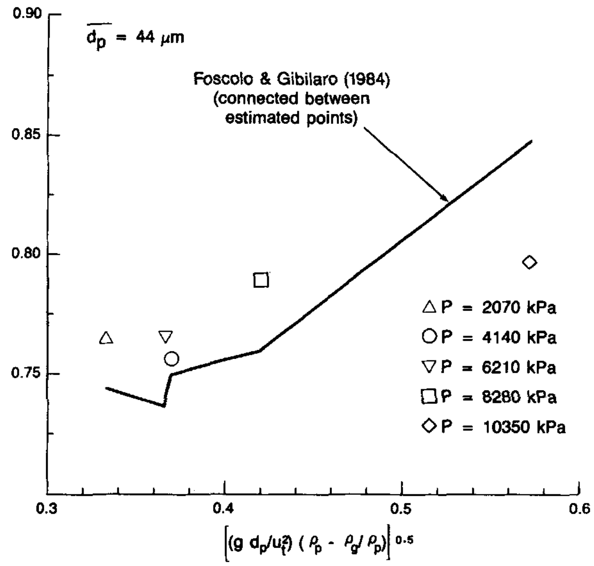


**Figure 9a. Experimental and estimated minimum bubbling voidage.**
Eq. 8 (F&G) with exp. $n$ and $u_t'$; $\bar{d}_p = 44\ \mu m$

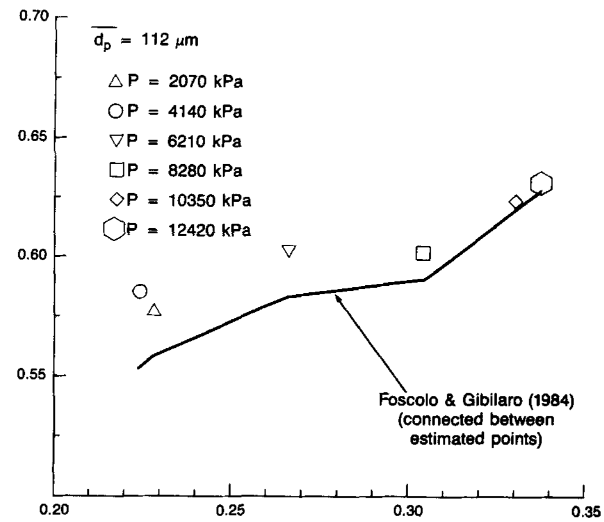


**Figure 9b. Experimental and estimated minimum bubbling voidage.**
Eq. 8 (F&G) with exp. $n$ and $u_t'$; $\bar{d}_p = 112\ \mu m$

powder. Correspondence for the 44 μm powder is poor due to the poor correspondence between $u_t'$ and $u_t$. These differences are probably due to the effect of large $F(\bar{d}_p = 44\ \mu m$ powder) on $u_t'$.

The validity of applying Eq. 8 *a priori* depends upon the validity of $u_t' = u_t$, and $n$ as calculated from the Richardson and Zaki formulas. Since experimental $n > 4.65$ and $u_t' > u_t$ were regarded as indicating the presence of interparticle forces (Geldart and Wong, 1984, among others), an appropriate theory for predicting $\epsilon_{mB}$ *a priori*, for systems of group A powders, may be one that is based upon the combination of interparticle (improved $n$ and $u_t'$ estimation) and hydrodynamic forces, eq. 8.

## Results and Conclusions

From the work reported here for group A carbon powders fluidized by synthesis gas ($M_W = 17$) within the pressure range $2{,}070 < P < 12{,}420$ kPa, it may be concluded that:

1. The expansion index $n$ decreases with increasing $P$ and, for all conditions, exceeds the values of Richardson and Zaki (1954).
2. Experimental values of $u_t'$, exceed calculated values of $u_t$ at all $P$ for the $\bar{d}_p = 44\ \mu m$ powder, but approximate closely those calculated for the $\bar{d}_p = 112\ \mu m$ powder.
3. $u_{mB}$ and $\epsilon_{mB}$ increase slightly with increasing $P$ for the 112 μm powder at all $P$, and for the 44 μm powder at all $P < 8{,}280$ kPa.
4. Above some $P$ within the range $8{,}280 < P < 10{,}350$ kPa for the 44 μm powder, $\epsilon$ increases immediately for $u_o > u_{mB}$; i.e., the apparent appearance of the first bubble is not accompanied by a decrease in total bed voidage. The minimum bubbling velocity $u_{mB}$ then appears to begin decreasing with increasing $P$.
5. Particulate bed expansion is adequately described by the equation of Foscolo et al. (1983), Eq. 5, for the 112 μm powder at all $P$.
6. Equation 5 underestimates particulate expansion for the 44 μm powder, most likely because this equation does not account for the effect of large $F$.
7. Equation 6, from Abrahamsen and Geldart (1980a), adequately describes particulate bed expansion for the 44 μm powder at all $P$.
8. Equation 7, from Abrahamsen and Geldart (1980b), adequately estimates $u_{mB}$ at all $P$ for the 44 μm powder.
9. The theory of Foscolo and Gibilaro (1984), Eq. 8, adequately estimates $\epsilon_{mB}$ for both powders at all $P$, provided that experimentally determined values of $n$ and $u_t'$ are applied in the theory.
10. Application of Eq. 8, utilizing experimentally determined values of $n$ and calculated values of $u_t$, adequately estimates $\epsilon_{mB}$ for the 112 μm powder at all $P$.
11. *A priori* application of Eq. 8 inaccurately predicts $\epsilon_{mB}$ for both powders at all $P$. This is because calculated values of $n$, taken from Richardson and Zaki (1954), are inadequate for the high-pressure systems studied here.

## Notation

$A_T$ = cross-sectional area of bed
$\bar{d}_p$ = mean particle diameter
$d_T$ = fluidized bed ID
$F$ = weight fraction of fine particles passing through a 45 μm sieve, i.e., $\bar{d}_p < 45\ \mu m$
$g$ = gravitational constant
$L_f$ = expanded bed height
$M_W$ = average gas molecular weight
$n$ = expansion index correlated via Eq. 1
$P$ = pressure
$Re_t = u_t\bar{d}_p\rho_g/\mu_g$, terminal Reynolds number
$Re_t' = u_t'\bar{d}_p\rho_g/\mu_g$, Reynolds number utilizing $u_t'$
$T$ = temperature
$u_o$ = superficial gas velocity
$u_{Do}$ = superficial dense phase gas velocity
$u_{mB}$ = minimum bubbling velocity
$u_{mf}$ = minimum fluidization velocity
$u_n$ = superficial gas velocity where normal bubbling commences
$u_t$ = particle terminal velocity
$u_t'$ = superficial gas velocity required to give $\epsilon = 1$
$W_T$ = total solids weight in bed

### *Greek letters*

$\Delta P$ = differential pressure
$\delta_B$ = dilute phase (bubble) volume fraction
$\delta_D$ = dense phase volume fraction
$\epsilon$ = overall bed voidage
$\epsilon_D$ = dense phase voidage
$\epsilon_{mb}$ = bed voidage at minimum bubbling
$\epsilon_{mf}$ = bed voidage at minimum fluidization
$\mu_g$ = gas viscosity
$\langle \rho_{bed} \rangle_c$ = measured cross-sectional centerline bed density
$\rho_g$ = gas density
$\rho_p$ = particle density
$\phi$ = particle sphericity

## Literature Cited


Abrahamsen, A. R., and D. Geldart, "Behavior of Gas-Fluidized Beds of Fine Powders. I: Homogeneous Expansion," *Powder Technol.*, **26**, 35 (1980a).

———, "Behavior of Gas-Fluidized Beds of Fine Powders. II: Voidage of the Dense Phase in Bubbling Beds," *Powder Technol.*, **26**, 47 (1980b).

Barreto, G. F., J. G. Yates, and P. N. Rowe, "The Effect of Pressure on the Flow of Gas in Fluidized Beds of Fine Particles," *Chem. Eng. Sci.*, **38**, 1935 (1983).

*Chemical Processing*, "Low Molecular Weight Alcohols Produced in Catalytic Process from CO and Hydrogen," p. 113 (Apr., 1985).

*Chemical Week*, "Getting Mixed Alcohols from Synthesis Gas," p. 28 (Nov. 7, 1984).

Chitester, D. C., R. M. Kornosky, L. S. Fan, and J. P. Danko, "Characteristics of Fluidization at High Pressure," *Chem. Eng. Sci.*, **39**, 253 (1984).

Clift, R., J. R. Grace, and M. E. Weber, *Bubbles, Drops, and Particles*, Academic Press, New York, 114 (1978).

Crowther, M. E., and J. C. Whitehead, "Fluidization of Fine Particles at Elevated Pressure," *Fluidization*, J. F. Davidson, D. L. Keairns, eds., Cambridge Univ. Press, 65 (1978).

De Jong, J. A. H., and J. F. Nomden, "Homogeneous Gas-Solid Fluidization," *Powder Technol.*, **9**, 91 (1974).

Ergun, S., "Fluid Flow Through Packed Columns," *Chem. Eng. Prog.*, **48**, 89 (1952).

Foscolo, P. U., and L. G. Gibilaro, "A Fully Predictive Criterion for the Transition Between Particulate and Aggregate Fluidization," *Chem. Eng. Sci.*, **39**, 1667 (1984).

Foscolo, P. U., L. G. Gibilaro, and S. P. Waldram, "A Unified Model for Particulate Expansion of Fluidized Beds and Flow in Fixed Porous Media," *Chem. Eng. Sci.*, **38**, 1251 (1983).

Geldart, D., "Types of Gas Fluidization," *Powder Technol.*, **7**, 285 (1973).

———, "Particle Entrainment and Carryover," *Gas Fluidization Technology*, D. Geldart, ed., Wiley, New York, 134 (1986).

Geldart, D., and A. C. Y. Wong, "Fluidization of Powders Showing Degrees of Cohesiveness. I: Bed Expansion," *Chem. Eng. Sci.*, **39**, 1481 (1984).

———, "Fluidization of Powders Showing Degrees of Cohesiveness. II: Experiments on Rates of Deaeration," *Chem. Eng. Sci.*, **40**, 653 (1985).

Gibilaro, L. G., R. Di Felice, S. P. Waldram, and P. U. Foscolo, "Generalized Friction Factor and Drag Coefficient Correlations for Fluid-Particle Interactions," *Chem. Eng. Sci.*, **40**, 1817 (1985).

Godard, K., and J. F. Richardson, "The Behavior of Bubble-Free Fluidized Beds," *Inst. Chem. Eng. Symp. Ser.*, **30**, 126 (1968).

Guedes de Carvalho, J. R. F., "Dense Phase Expansion in Fluidized Beds of Fine Particles," *Chem. Eng. Sci.*, **36**, 413 (1981).

Guedes de Carvalho, J. R. F., D. F. King, and D. Harrison, "Fluidization of Fine Particles Under Pressure," *Fluidization*, J. F. Davidson, D. L. Keairns, eds., Cambridge Univ. Press, Cambridge, 59 (1978).

Juhola, A. J., and E. O. Wiig, "Pore Structure in Activated Charcoal. I: Determination of Micropore Size Distribution," *J. Am. Chem. Soc.*, **71**, 2069 (1949).

King, D. F., and D. Harrison, "The Dense Phase of a Fluidized Bed at Elevated Pressures," *Trans. Inst. Chem. Eng.*, **60**, 26 (1982).

Knight, M. J., and P. N. Rowe, "On Measuring the Density of Fine Particles by Pycnometry," *Chem. Eng. Sci.*, **35**, 997 (1980).

Knight, M. J., P. N. Rowe, H. J. MacGillivray, and D. J. Cheesman, "On Measuring the Density of Finely Divided but Porous Particles such as Fluid Bed Cracking Catalyst," *Trans. Inst. Chem. Engr.*, **58**, 203 (1980).

Lockett, M. J., and G. Gunnarsson, "The Delayed Bubbling of Gas-Fluidized Beds," *Chem. Eng. Sci.*, **28**, 666 (1973).

Massimilla, L., G. Donsi, and C. Zucchini, "The Structure of Bubble-Free Gas-Fluidized Beds of Fine Fluid Cracking Catalyst Particles," *Chem. Eng. Sci.*, **27**, 2005 (1972).

Murchison, C. B., "The Use of Molybdenum Catalysts for Fischer-Tropsch," *Proc. Climax 4th Int. Conf. Chem. Uses Molybdenum*, H. F. Barry, P. C. H. Mitchell, eds., Climax Molybdenum Co., Ann Arbor, MI, 197 (1982).

Piepers, H. W., E. J. E. Cottaar, A. H. M. Verkooijen, and K. Rietema, "Effects of Pressure and Type of Gas on Particle-Particle Interaction and the Consequence for Gas-Solid Fluidization Behavior," *Powder Technol.*, **37**, 55 (1984).

Richardson, J. F., "Incipient Fluidization and Particulate Systems," *Fluidization*, J. F. Davidson, D. Harrison, eds., Academic Press, London, 26 (1971).

Richardson, J. F. and W. N. Zaki, "Sedimentation and Fluidization," *Trans. Inst. Chem. Engr.*, **32**, 35 (1954).

Rietema, K., "The Effect of Interparticle Forces on the Expansion of a Homogeneous Gas-Fluidized Bed," *Chem. Eng. Sci.*, **28**, 1493 (1973).

Rowe, P. N., P. U. Foscolo, A. C. Hoffman, and J. G. Yates, "Fine Powders Fluidized up to 20 Bar with Gases of Different Viscosity," *Chem. Eng. Sci.*, **37**, 1115 (1982).

Sciazko, M., and J. Bandrowski, "Effect of Pressure on the Minimum Bubbling Velocity of Polydisperse Materials," *Chem. Eng. Sci.*, **40**, 1861 (1985).

Simone, S., and P. Harriott, "Fluidization of Fine Powders with Air in the Particulate and the Bubbling Regions," *Powder Technol.*, **26**, 161 (1980).

Sobreiro, L. E. L., and J. L. F. Monteiro, "The Effect of Pressure on Fluidized Bed Behavior," *Powder Technol.*, **33**, 95 (1982).

Toomey, R. D. and H. F. Johnstone, "Gaseous Fluidization of Solid Particles," *Chem. Eng. Prog.*, **48**, 221 (1952).

Wallis, G. B., *One-Dimensional Two-Phase Flow*, McGraw-Hill, New York (1969).

Weimer, A. W., and K. V. Jacob, "On Bed Voidage and Apparent Dilute Phase Holdup in High-Pressure Turbulent Fluidized Beds of Fine Powders," *Fluidization V*, K. Ostergaard, A. Sorensen, eds., Eng. Found., New York, 313 (1986a).

———, "Bed-to-Surface Heat Transfer for Vertical U-Tubes in a High-Pressure Slugging Hydrogen-Fluidized Bed of Carbon-Supported Catalyst Powder," *Powder Technol.*, **48**, 247 (1986b).

Weimer, A. W., and G. J. Quarderer, "Effect of Temperature on the Dense Phase in High-Pressure Fluidized Beds of Fine Powders," *AIChE Symp. Ser., No. 241*, **80**, 79 (1984).

———, "On Dense Phase Voidage and Bubble Size in High-Pressure Fluidized Beds of Fine Powders," *AIChE J.*, **31**, 1019 (1985).

Weimer, A. W., D. C. Gyure, and D. E. Clough, "Application of a Gamma-Radiation Density Gauge for Determining Hydrodynamic Properties of Fluidized Beds," *Powder Technol.*, **44**, 179 (1985).


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