[*This manuscript is dedicated to the memory of C. Y. Wen*]

Gas Entry Effects in Fluidized Bed Reactors

Experimental results from an X-ray study of gas bubbles entering a fluidized bed of two differently sized powders are presented and their significance for chemical reactions taking place in the distributor region of a fluidized bed reactor are examined using a simple two-phase model.

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SCOPE

It is a widely accepted experimental fact from both industrial and laboratory experiments that a disproportionate amount of chemical reaction occurs in the bottom few centimeters of a gas fluidized bed chemical reactor. The effect is particularly noticeable with fast reactions, and it seems fairly obvious that mass transfer is good in this shallow region and far less good elsewhere in the bed. It has been known for a long time that under conditions that commonly obtain in a reactor, the bubbles are a major cause of bypassing or poor gas/solid mass transfer (Yates, 1983). Normally bubbles originate from the holes or other orifices in the distributor plate on which the bed rests and it is reasonable to suppose that good contacting and mass transfer are associated with the process of bubble formation.

The mechanism by which gas enters a fluidized bed and subsequently forms bubbles is not easy to observe, and theoretical understanding of the fluidized system is inadequate to propose a mechanism. The major experimental difficulty is that, unlike gases in inert liquids, the bed is opaque and the bubbles cannot normally be seen. The use of "two-dimensional" beds or the introduction of transparent surfaces distorts the hydrodynamics and can produce misleading results. X-ray ciné photographs cause no distortion; although the method has limitations, it does give valuable information on the formation process.

Results are described in this paper and then used to formulate a model that gives a plausible and semiquantitative explanation of the observed enhanced reaction rate.

CONCLUSIONS AND SIGNIFICANCE

X-ray examination of the distributor region of a fluidized bed of alumina particles shows that for the range of conditions investigated, gas entering the bed from a centrally located orifice does so in the form of bubbles which form at a constant frequency of about 7 Hz. The gas flow visible as bubbles is only about one third of that entering the bed; it is assumed that the rest penetrates the dense phase around the bubble and causes its voidage to increase above the minimum fluidization value. At a distance of 25 cm above the distributor, bubble frequency has decreased to about 4 Hz and some three quarters of the entering gas is visible as bubbles. The implications of this gas entry mechanism for chemical reactions in the region of the distributor are discussed in terms of a simple model; it is shown that for fast reactions a considerable amount of conversion can be achieved in the zone close to the point of gas entry.

INTRODUCTION

About two decades ago Davidson and Schüler (1960) studied theoretically the entry of gas from an orifice into both viscous and inviscid liquids. They postulated that gas enters as a spherical bubble which, as it forms, rises in the fluid due to the action of bouyancy forces and detaches from the orifice when its rate of rise exceeds its rate of radial growth (Figure 1).

The bubble volume is given by:

$$V_b = \frac{4}{3}\pi R^3 = Q_{or}\, t \tag{1}$$

where Q_{or} is the orifice gas flow rate.

Now

$$\frac{dt}{dR} = \frac{4}{3}\frac{\pi}{Q_{or}} 3R^2 \tag{2}$$

and hence

$$\frac{dR}{dt} = \frac{Q_{or}}{4\pi R^2} \tag{3}$$

The rate of rise of the bubble (Davies and Taylor, 1950) is

$$\frac{dx}{dt} = K\sqrt{gR} \tag{4}$$

and detachment occurs when:

$$\frac{dx}{dt} \geqslant \frac{dR}{dt}$$

i.e., when

$$K\sqrt{gR} \geqslant \frac{Q_{or}}{4\pi R^2} \tag{5}$$

From Eq. 5 the minimum bubble radius at the point of detachment is:

$$R_1 \left(\frac{Q_{or}}{4\pi K\sqrt{g}}\right)^{2/5} \tag{6}$$

from which the initial, spherical, bubble volume is clearly:

$$V_{b1} = \frac{4}{3}\pi\left[\left(\frac{Q_{or}}{4\pi K\sqrt{g}}\right)^{2/5}\right]^3 = K'\frac{Q_{or}^{1.2}}{g^{0.6}} \tag{7}$$

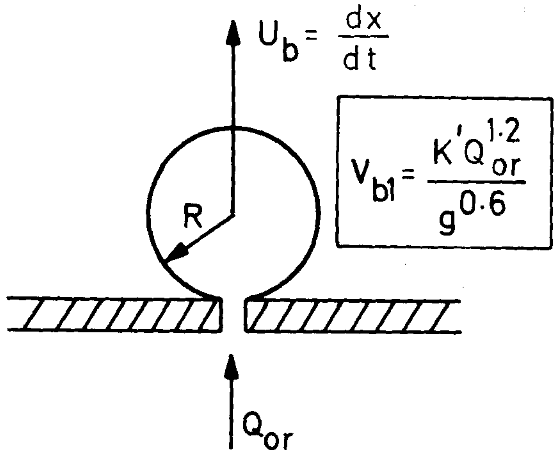

Figure 1. Davidson-Schüler model of gas entering a liquid from an orifice.

Davidson and Schüler studied experimentally the entry of air into water using high-speed photography and found that bubbles do in fact form in the way postulated and that at low gas flow rates the above equation is obeyed quite well. However, it is no longer true at high flow rates where the bubble shape is distorted by turbulence in the liquid and by inertial effects. Subsequently, Harrison and Leung (1961) carried out a similar study using a gas fluidized bed in place of water. The gas was observed to enter in the same way, as a stream of bubbles, the frequencies of formation of which were measured by means of a capacitance probe fitted inside the orifice from which the gas was discharged. They then calculated initial bubble volumes from this measured frequency and the inlet flow rate and concluded that the Davidson-Schüler equation fitted their data well at low flow rates. The bubble frequencies they measured at high flow rates (600–8000 cm^3/s through a 2.54 cm diameter orifice) were in the range 18–21 Hz and they calculated the gas leakage through the roof of a bubble during its formation to be no more than 15%.

Nguyen and Leung (1972) injected air through an orifice into an incipiently fluidized two-dimensional bed of alumina particles and found considerable leakage of gas into the emulsion phase during the process of bubble formation: they correlated their observed bubble volumes with inlet gas flow rate and frequency of bubble formation, n_b, as:

$$V_{b1} = 0.53 \frac{Q_{or}}{n_b} \tag{8}$$

Nguyen and Leung also found the bubble formation frequency to vary with increasing Q_{or} in accordance with the Davidson-Schüler equation; the observed lower limit of n_b was about 10 Hz at Q_{or} = 100 cm^3/s.

Rowe et al. (1979) used X-ray ciné photography to investigate the entry of gas from an orifice into a fluidized bed with the object of establishing whether bubbles or stable jets were produced. They observed nothing but bubbles in a bed 15 cm in diameter fitted with orifices of 6.4–15.9 mm diameter and at gas velocities in the range 1–70 m/s. They also measured a bubble formation frequency of 8 Hz and found it to be constant over the whole range of velocities. Subsequent analysis of their film showed the visible bubble flow on entry to be less than the total flow through the orifice by up to a factor of two; only about half the gas flowing into the bed as bubbles actually appeared as bubbles at the orifice. Some distance above the grid, however, the volumetric flows were more or less in balance.

Wen et al. (1982) also set out to establish criteria for the formation of either jets or bubbles at the distributor of a three-dimensional bed. They concluded that jets are formed only when the gas orifice is surrounded by stagnant zones of particles.

A recent publication from Westinghouse (Yang et al., 1983) reported studies on a 3 m diameter semicylindrical bed in which, as with the two-dimensional work of Nguyen and Leung (1972), bubbles and jets could be observed directly against the plane wall of the vessel. Yang et al. concluded that a substantial amount of gas leaks from the bubble to the emulsion phase during bubble formation particularly when the bed is less than minimally fluidized. They gave correlations for two sets of operating conditions in the same form as Eq. 8 with values of the coefficient on the righthand side of the equation of 0.303 and 0.118.

The work to be reported here is a continuation of the previously mentioned X-ray study of Rowe et al. (1979) and was carried out to determine more precisely the fraction of gas that leaks from a bubble during its formation at an orifice.

EXPERIMENTAL

An aluminium vessel of rectangular cross section (20 cm × 30 cm) was set up in the X-ray unit described in detail by Rowe et al. (1979). It was fitted with a porous plate distributor containing a central orifice of diameter 1 cm; the powder in the bed was held at its point of minimum fluidization and air at a flow rate, Q_{or}, in the range 67–2,100 cm^3/s (0.85–26.75 m/s) was admitted through the orifice. Two powders were studied: (i) granular alumina in the size range +200–400 μm with a U_{mf} value of 2.94 cm/s (powder 1); (ii) granular alumina of size −150 μm and of U_{mf} 0.21 cm/s (powder 2).

Ciné films of the bubbling bed were taken at 42 frames/s at two positions, 10 and 25 cm above the distributor. With the camera in the lower position events occurring immediately above the distributor were visible. Bubble frequencies at both positions (n_{b1} and n_{b2}) were obtained directly from the films while bubble volumes (V_{b1} and V_{b2}) were measured by means of a Hewlett-Packard 9874 A digitiser coupled to a Commodore 4032 computer (Rowe et al., 1983). At least 100 bubbles were analyzed for each set of operating conditions and average values of frequencies and volumes were determined.

RESULTS

These are given in Table 1 from which it is immediately clear that the frequency of bubble formation at the orifice remained remarkably constant over the whole range of flow rates studied; the mean values for n_{b1} for powders 1 and 2 were 6.82 and 6.93 Hz, respectively. Furthermore, it is apparent that 25 cm above the distributor bubble frequencies had fallen to about half their value at entry, and bubble volumes, V_{b2}, had increased considerably, both as the result mainly of coalescence.

A comparison between the flow through the orifice and the visible bubble flow at both positions is shown in Figures 2 and 3 where V_b is plotted against Q_{or}/n_b for the two powders. The relationships are reasonably linear and are represented by the two equations:

$$V_{b1} = 0.36 Q_{or}/n_{b1} \tag{9}$$

$$V_{b2} = 0.79 Q_{or}/n_{b2} \tag{10}$$

From Eq. 9 it would appear that only about one third of the gas entering the bed at the orifice actually appears as a bubble with a well-defined periphery, the balance presumably penetrating the emulsion phase and causing its local voidage to increase above that

TABLE 1. OBSERVED BUBBLE VOLUMES AND FREQUENCIES

Powder	Q_{or} (cm^3/s)	V_{b1} (cm^3)	n_{b1} (Hz)	V_{b2} (cm^3)	n_{b2} (Hz)
1	233	12.4	6.07	29.7	3.16
1	700	35.7	6.43	167	2.97
1	1,400	56.0	7.54	306	3.36
1	2,100	112	7.23	460	3.66
2	67	4.6	6.20	8.4	4.01
2	200	17.3	6.84	50.8	3.75
2	400	26.5	7.20	75.8	4.25
2	600	28.7	7.49	77.0	5.31

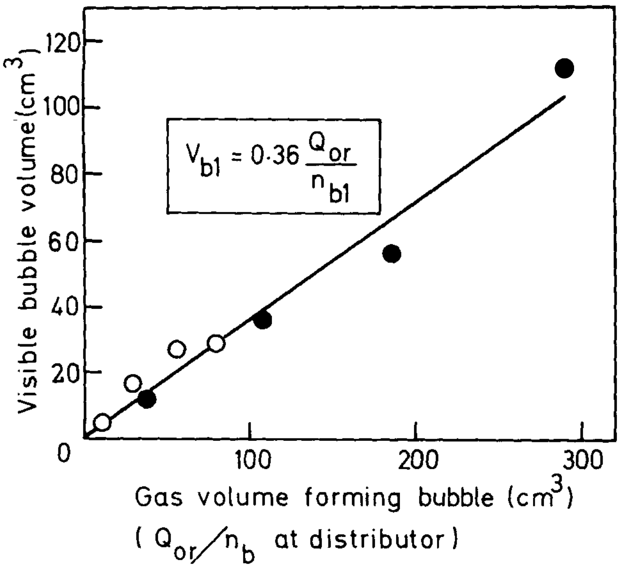

Figure 2. Volume of visible bubble, V_{b1}, at point of detachment from an orifice as a function of gas flow through the orifice divided by bubble frequency, Q_{or}/n_{b1};

● coarse alumina powder

○ fine alumina powder

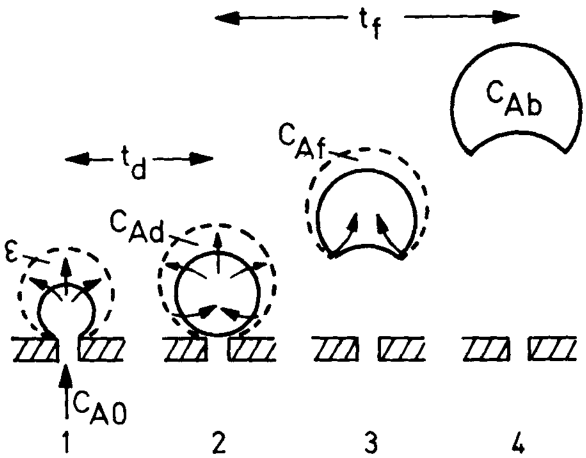

Figure 4. Proposed model of gas entering a fluidized bed from an orifice.

1. bubble boundary grows by gas flowing past the particles and so exerting a drag force on them.
2. bubble detaches and rises into a region of voldage greater than ϵ_{mf} (unstable).
3. excess gas flows back into the bubble allowing particles to return to stable voldage ϵ_{mf}.
4. stable bubble bigger than size at detachment established some distance above orifice.

at minimum fluidization (Figure 4). As the bubble rises in the bed some of this "invisible" gas flows from the emulsion into the bubble void so that at a distance of 25 cm above the distributor some three quarters of the entering gas is accounted for in the bubble phase. Such high rates of gas leakage from a bubble during its formation at an orifice can thus lead to considerable deviations from the ideal two-phase theory of fluidization particularly in the lower regions of the bed and it is conceivable that in some instances a single bubble will leave the bed surface before it has attained its equilibrium size.

The results of our observations are in broad agreement with those mentioned earlier of Nguyen and Leung (1972) and of Yang et al. (1983) made in two-dimensional and semicylindrical beds, respectively. At low gas flow rates near spherical bubbles rise periodically from the orifice at the characteristic frequency of about 7 Hz. Coalescence between succeeding bubbles occurs from time to time several centimeters above presumably as a following bubble accelerates in the particle drift behind its predecessor. With increasing gas flow rate coalescence occurs more frequently and closer to the origin and increasingly three, four or even five successive bubbles coalesce almost immediately on entering. However, even at the highest flow rates observed when rapid and immediate coalescence is the rule, detailed examination of the individual frames on the film show distinct separate bubble formation without exception. That is to say, there is always some period during the 7 Hz cycle when the hole is covered by particles. Even in these very disturbed conditions each entering bubble shows a reasonably well shaped spherical upper surface before distorting on coalescence. After one of these bursts of up to five bubbles coalescing almost immediately there is a pause as another burst accumulates so that at a few centimeters above a fairly regular stream of quite well shaped bubbles passes at a much lower frequency than 7 Hz.

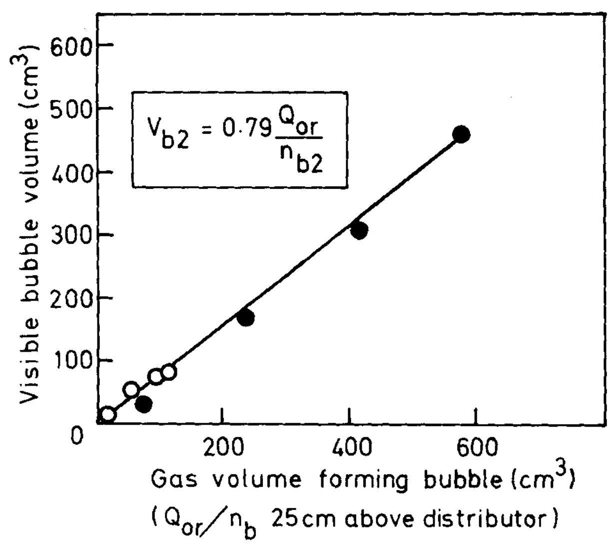

Figure 3. Volume of visible bubble, V_{b2}, at a point 25 cm above an orifice as a function of gas flow through the orifice divided by bubble frequency Q_{or}/n_{b2};

● coarse alumina powder

○ fine alumina powder

DISCUSSION

Our work on the mode of gas entry from an orifice into a fluidized bed is still at an early stage and our current views may be modified by further studies. Nonetheless, it is worth speculating on the effect the mechanism presented above could have on the course of a chemical reaction in a fluidized bed. Let us then assume that during the period up to the time of detachment of the bubble, t_d, the unaccounted or invisible gas has passed into the dense phase causing the particles to expand to a void fraction, ϵ, in excess of ϵ_{mf} (Figure 4). The time t_d is clearly related to the frequency of bubble formation n_b by:

$$t_d = \frac{1}{n_b} \tag{11}$$

and during this period the volume of gas entering the dense phase and reacting is $Q_{or}t_d\,(1 - x_o)$ where x_o is the fraction of flow that forms the visible bubble. Further if we assume the gas entering the dense phase undergoes a pseudohomogeneous first-order reaction with the particles at a rate given by:

$$-r_A = (1 - \epsilon)k_sC_A \tag{12}$$

where k_s is the first-order rate constant, then the fraction of reactant A remaining unconverted after time t is:

$$\frac{C_A}{C_{ao}} = \exp[-(1-\epsilon)k_s t]. \quad (13)$$

Now if the time taken for the bubble gas to reach equilibrium (i.e., the time for the bubble to form completely) is t_e and if the mean residence time of the gas in the expanded dense phase is 0.5 t_e, then Eq. 13 becomes:

$$\frac{C_A}{C_{Ao}} = \exp[-0.5(1-\epsilon)k_s t_e]. \quad (14)$$

When the bubble has formed completely the gas it contains will be made up partly of that which has been in contact with the dense phase and partly of that which has not, so that at equilibrium the fraction of A remaining unreacted inside the bubble will be:

$$\frac{C_{Ab}}{C_{Ao}} = \left\{\frac{Q_{or}t_d x_o + Q_{or}t_d(1-x_o)\dfrac{C_A}{C_{Ao}}}{Q_{or}\,t_d}\right]$$

$$= x_o + (1-x_o)\exp[-0.5(1-\epsilon)k_s t_e] \quad (15)$$

Assuming a value of the rate constant k_s is available from auxiliary experiments in, for example, a fixed bed reactor, the bubble-phase concentration of reactant gas may be calculated once the three remaining parameters x_o, ϵ, and t_e are known.

In the present work the proportion of incoming gas appearing as a visible bubble at the orifice (x_o = 0.36) and at a position 25 cm above the orifice (x_{25} = 0.79) have been measured and so by a linear extrapolation from these two values the height at which x = 1.0, and hence at which the bubble gas has reached equilibrium, may be found. If this height is H_e then the time for the bubble to reach equilibrium is clearly:

$$t_e = \frac{H_e}{U_b} \quad (16)$$

so that if the bubble velocity is known the residence time of gas in the expanded dense phase may be calculated. Now the velocity of a bubble in a fluidized bed is normally expressed in terms of its diameter, d_b, by the Davies-Taylor relationship:

$$U_b = \left(\frac{gd_b}{2}\right)^{1/2} \quad (17)$$

the observed coefficient of proportionality being unity. As approximately one quarter of the volume of the sphere centered on the bubble is occupied by bubble wake, the volume of gas forming the bubble is:

$$V_b = \frac{3}{4}\frac{\pi}{6}d_b^3. \quad (18)$$

$$= x_o\frac{Q_{or}}{n_b} \quad (18a)$$

Hence:

$$d_b = \left(\frac{8}{\pi}\right)^{1/3} V_b^{1/3}, \quad (19)$$

$$U_b = \left(\frac{g}{2}\right)^{1/2}\left(\frac{8}{\pi}\right)^{1/6} V_b^{1/6}$$

$$= 0.259\, V_b^{1/6} \quad (20)$$

and combining Eqs. 16, 18a and 20:

$$t_e = 3.86\, H_e\left(\frac{n_b}{Q_{or}}\right)^{1/6} \quad (21)$$

The value of the third parameter in Eq. 15, the voidage of the expanded dense phase, ϵ, is at present unknown but reasonable values in the range 0.4–0.7 may be assumed for the purpose of an illustrative calculation. Combining Eqs. 15 and 21 the fraction of reactant A remaining unreacted in the bubble phase at a height H_e above the distributor is:

TABLE 2. CALCULATED FRACTION UNCONVERTED LEAVING DISTRIBUTOR ZONE

	C_{Ab}/C_{Ao}		
ϵ	$k_s = 1\ s^{-1}$	$k_s = 5\ s^{-1}$	$k_s = 10\ s^{-1}$
0.40	0.876	0.577	0.434
0.50	0.895	0.620	0.466
0.60	0.914	0.672	0.512
0.70	0.934	0.733	0.577

$$\frac{C_{Ab}}{C_{Ao}} = x_o + (1-x_o)\exp\left[-1.93(1-\epsilon)k_s H_e\left(\frac{n_b}{Q_{or}}\right)^{1/6}\right] \quad (22)$$

Example

The value of H_e calculated from the foregoing measurements of visible bubble volume at two bed heights (0 and 0.25 m above the orifice) is 0.375 m. Other values used in the calculation are as follows:

$$x_o = 0.36$$

$$n_b = 7\ \text{Hz}$$

$$Q_{or} = 5 \times 10^{-4}\ \text{m}^3/\text{s}$$

$$k_s = 1, 5, 10\ \text{s}^{-1}$$

$$\epsilon = 0.4, 0.5, 0.6, 0.7$$

Table 2 gives the calculated values of C_{Ab}/C_{Ao} at height H_e above the distributor and it is clear that, particularly in the case of fast reactions, the model predicts considerable conversion in the distributor zone.

CONCLUDING REMARKS

This model is quite simple in conception and rests on the mechanism of bubble formation that has been observed over a range of conditions. It certainly offers a plausible qualitative explanation for the commonly observed behavior of real reactors, but as yet there is insufficient data critically to test the model. The model supposes that a large part of the gas which ultimately forms a bubble with restricted mass transfer to the particles first enters the dense phase for a brief period during which appreciable chemical reactions may occur if it is a fast reaction. The few parameters that as yet are unknown or imperfectly known are capable of measurement by physical methods which would then allow calculation from first principles in particular cases.

LITERATURE CITED

Davidson, J. F., and B. O. G. Schüler, "Bubble Formation at an Orifice in an Inviscid Liquid," *Trans. Instn. Chem. Engrs.*, **38**, 335 (1960).

Davies, R. M., and G. I. Taylor, "The Mechanics of Large Bubbles Rising Through Extended Liquids and Through Liquids in Tubes," *Proc. Roy. Soc.*, **A200**, 375 (1950).

Harrison, D., and L. S. Leung, "Bubble Formation at an Orifice in a Fluidised Bed," *Trans. Instn. Chem. Engrs.*, **39**, 409 (1961).

Nguyen, X. T., and L. S. Leung, "A Note on Bubble Formation at an Orifice in a Fluidized Bed," *Chem. Eng. Sci.*, **27**, 1748 (1972).

Rowe, P. N., H. J. MacGillivray, and D. J. Cheesman, "Gas Discharge from an Orifice into a Gas Fluidized Bed," *Trans. Instn. Chem. Engrs.*, **57**, 194 (1979).

Wen, C. Y., N. R. Deole, and L. H. Chen, "A Study of Jets in a Three-Dimensional Gas Fluidized Bed," *Powder Tech.*, **31**, 175 (1982).

Yang, W., et al., "Fluidization Phenomena in a Large Scale, Cold Flow Model," *Fluidization IV*, D. Kunii and R. Toei, Eds., Plenum Press, New York, 77 (1983).

Yates, J. G., *Fundamentals of Fluidized-Bed Chemical Processes*, Butterworths, London (1983).

NOTATION

d_b = bubble diameter, m
C_a = concentration of reactant A, mol/m^3
g = acceleration due to gravity, m/s^2
H_e = height at which bubble is fully formed, m
k_s = reaction rate constant from packed bed data, s^{-1}
n_b = bubble frequency, Hz
Q = volumetric gas flow rate into expanded dense phase, m^3/s
Q_{or} = volumetric gas flow rate through orifice, m^3/s
R = bubble radius, m
r_A = rate of reaction of A, mol/m^3·s
t_d = bubble formation time, s
t_e = bubble equilibration time, s
U_b = bubble rise velocity, m/s
U_{mf} = minimum fluidization velocity, m/s
V_b = visible bubble volume, m^3
x = fraction of orifice flow forming a visible bubble
ϵ = dense-phase voidage
ϵ_{mf} = dense-phase voidage at minimum fluidization

Subscripts

1 = as seen 10 cm above distributor
2 = as seen 25 cm above distributor
0 = entry condition
d = dense phase

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Distribution of Bubble Properties in a Gas-Liquid-Solid Fluidized Bed

The behavior of bubbles in a cocurrent gas-liquid-solid fluidized bed was investigated in a column of 76.2 mm ID in this study. The particles used were glass beads of 3 and 6 mm and a binary mixture of these particles. A novel dual electrical resistivity probe system was developed and utilized to obtain bubble properties including bubble size and rise velocity. The distributions of the bubble properties in the gas-liquid-solid fluidized bed were evaluated for three flow regimes: the dispersed bubble flow regime; the coalesced bubble flow regime; and the slug flow regime.

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SCOPE

Gas-liquid-solid fluidized beds have been fully developed and demonstrated in processing technology; as three-phase reactors, they have been employed in the H-oil process for hydrogenation and hydrodesulfurization of residual oil, the H-coal process for coal liquefaction, and the bio-oxidation process for waste water treatment.

A knowledge of hydrodynamics is of considerable importance in design and operation of a gas-liquid-solid fluidized bed. Among various hydrodynamic properties, bubble size and rise velocity and their distributions are of prime concern, as they are directly responsible for the behavior of other hydrodynamic properties such as liquid flow patterns, solids mixing, and gas-liquid interfacial area.

Very limited information is available in the literature regarding the in-bed bubble size or rise velocity distributions in the gas-liquid-solid fluidized bed. Page and Harrison (1972) used photographic techniques to examine the size distribution of bubbles leaving a gas-liquid-solid fluidized bed and found that the logarithmic cumulative distribution function varies linearly with bubble size. Using an impedance double-probe, Darton and Harrison (1974) reported that bubble size in a three-phase fluidized bed of 500 μm sand particles follows a log-normal distribution.

This study is directed toward a fundamental understanding of in-bed bubble properties including bubble size and bubble rise velocity in a gas-liquid-solid fluidized bed of large particles and their mixture. The bubble properties are distinguished for each of the three flow regimes; dispersed bubble flow regime; coalesced bubble flow regime; and slug flow regime.

CONCLUSIONS AND SIGNIFICANCE

A novel dual electrical resistivity probe was developed to obtain the distribution of bubble rise velocity and size in a gas-liquid-solid fluidized bed containing large particles. The probe used possessed adequate strength to sustain the impact of solid particles, yet was small enough to allow bubbles of small size to be detected. The experiments were conducted in a circular plexiglas column of 76.2 mm ID. The particles used were 3 and 6 mm glass beads and a binary mixture of them. The behavior of bubble properties in each of the three flow regimes

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