

0009-2509(94)00274-6

A UNIFIED APPROACH TO THE SCALE-UP OF GAS-SOLID FLUIDIZED BED AND GAS-LIQUID BUBBLE COLUMN REACTORS

J. ELLENBERGER and R. KRISHNA*

Department of Chemical Engineering, University of Amsterdam, Nicuwe Achtergracht 166, 1018 WV Amsterdam, The Netherlands

(Received 9 May 1994; accepted for publication 12 September 1994)

Abstract—The aim of this work is to develop a unified approach to the scale-up of gas-solid (G-S) fluid bed and gas-liquid (G-L) bubble column reactors. The unified approach relies on analogies in the hydrodynamic behavior of G-S and G-L systems in both the homogeneous and heterogeneous flow regimes. The homogeneous G-S fluidization regime is to be identified with homogeneous G-L bubbly flow while the heterogeneous G-S fluidization regime is to be identified with the churn-turbulent regime for G-L bubble columns. In the heterogeneous flow regime of operation, the classic two-phase theory developed for G-S fluidized beds can be applied with profit to describe also the hydrodynamics of G-L bubble columns provided the "dilute" phase is identified with the fast-rising large bubbles, and the "dense" phase is identified with the liquid phase containing entrained "small" bubbles.

Quantitative analogies in the hydrodynamic behavior of G-S and G-L systems are demonstrated by the use of extensive experimental data obtained in five columns of three different diameters $(2 \times 0.1 \text{ m}, 1 \times 0.19 \text{ m} \text{ and } 2 \times 0.38 \text{ m})$. The total expanded bed height in the experiments varied in the range 0.5-3.5 m. About 4,000 dynamic gas disengagement experiments were carried out with various systems to determine the total gas hold-up and the hold-ups of the "dilute" and "dense" phases. The gas phases used in the experiments were air, helium, argon and sulfur hexafluoride. Fluidized cracking catalyst (FCC) was used as the solid phase in fluid bed experiments. In bubble column operations the liquid phase used was water, paraffin oil or tetradecane. Sintered plates were used for gas distribution in all the columns.

The gas hold-up in the "dense" phase, e_{df} was found to be practically independent of the scale of operation. The hold-up of the fast-rising "dilute" phase, e_b , on the other hand was found to be a significant function of the column diameter, D_T , and of the total dispersion height, H. The "dilute" phase gas hold-up can be modeled for both G-S and G-L systems using a theory allowing for bubble growth in the region above distributor. The bubbles are assumed to grow in diameter up to a distance h^* , at which the bubbles reach their equilibrium size. The equilibration height h^* was found to increase with the superficial gas velocity through the dilute phase, $(U - U_{df})$. For air-FCC, the value of h^* varies in the range 0.4–1.2 m. For bubble columns the values of h^* are significantly smaller and lie in the range 0–0.5 m.

Increasing gas density increases the total gas voidage in both G–S and G–L systems but has no significant effect on the hold-up of the dilute phase. In G–L bubble columns, the liquid properties affect the total gas hold-up but have only a minor influence on the dilute phase hold-up.

The unified model to describe the bubble hydrodynamics in G-S fluid beds and G-L bubble columns is a useful tool in scaling-up these two reactor types, because of the possibilities of cross-fertilization of design data,

INTRODUCTION

An important key to the scale-up of gas-solid (G-S) fluid bed and gas-liquid (G-L) bubble column reactors is the proper description of the bubble hydrodynamics in these systems. It is generally appreciated that the performance of G-S fluid beds is a strong function of the scale of operation and that the column diameter and bed height have a profound influence on the bubble hold-up, interphase mass transfer and backmixing (Werther, 1983; Van Swaaij, 1985; Krishna, 1993). It is also well known that G-L bubble columns of diameter

smaller than about 0.05 m tend to slug when the superficial gas velocity exceeds about 0.1 m/s (Deckwer, 1992). For non-slugging operations, the column diameter has been found to influence the gas hold-up (Schumpe and Grund, 1986; Grund et al., 1992). While several analogies in the hydrodynamics of G-S fluid beds and G-L bubble columns have been pointed out in the published literature (Davidson et al., 1977; Miyauchi et al., 1981; Krishna, 1993; Krishna et al., 1993), a unified quantitative picture of the hydrodynamics is still to emerge. This paper aims at developing such a unified and quantitative approach to the scale-up of these two important types of reactors. The development is begun with a short introduction to flow regimes and flow regime transitions.

^{*}To whom correspondence should be addressed.

When a gaseous phase is introduced uniformly through the bottom of a packed bed of particles [see Fig. 1(a)] the bed begins to expand for gas velocities exceeding the minimum fluidization velocity U_{mf} . For fine particles, say smaller than 200 μ m, the bed expands uniformly; this is the regime of homogeneous fluidization. This regime of homogeneous fluidization prevails until a certain velocity is reached at which bubbles are first observed; the velocity at this point, U_{mb} , is usually called the minimum bubbling velocity. For the purposes of drawing analogies with gas-liquid systems, this velocity will be denoted as the transition velocity, U_{trans} . The operating gas velocity window between U_{mf} and U_{trans} is usually very narrow and it is usually not possible to operate commercial reactors in a stable manner in this regime. On the other hand, in gas-solid beds of large particles, say larger than 1 mm, bubbles appear as soon as the gas velocity exceeds U_{mf} and hence $U_{trans} \cong U_{mf}$. Beyond the gas velocity corresponding to U_{trans} is the regime of heterogeneous fluidization. In the heterogeneous fluidization regime, a small portion of the entering gas is used to keep the solids in suspension while the major portion of the gas flows through the reactor in the form of bubbles. Commercial reactors usually operate in the heterogeneous or bubbling fluidization regime as gas velocities U exceeding 0.1 m/s, a few orders of magnitude higher than U_{trans} . Under these conditions, the bubbles tend to rise up the column very quickly, and virtually in plug flow, at velocities of the order of 1.5 m/s, "by-passing" the suspended particles. These bubbles tend to churn up the bed causing the solid phase to be thoroughly backmixed. For highly exothermic reactions, such as regeneration of coked catalyst in Fluid Catalytic Cracking (FCC) regenerators, this backmixing characteristic is desirable from the point of view of thermal equilibration of the reactor contents.

An analogous picture emerges if one sparges gas into a column filled with a liquid; see Fig. 1(b). The bed of liquid begins to expand as soon as gas is introduced. Therefore, if U_{mf} is defined as the minimum fluidization velocity for a gas-liquid system it can be seen that $U_{mf} = 0$. As the gas velocity is increased beyond the value U_{mf} , the bed of liquid expands homogeneously and the bed height increases almost linearly with the superficial gas velocity. This regime of operation of a bubble column is called the homogeneous bubbly flow regime; this regime is analogous to the regime of homogeneous fluidization for a gas-solid system. The bubble size distribution is narrow and a roughly uniform bubble size, in the range 2-7 mm, is found. As the gas velocity is increased beyond U_{mf} , the bubble population increases and at a certain gas velocity, U_{trans} , coalescence of the bubbles takes place to produce the first fast-rising "large" bubble. The appearance of the first large bubble changes the hydrodynamic picture dramatically. The regime of operation for superficial gas velocities exceeding U_{trans} is commonly referred to as *heterogeneous* or *churn-turbulent regime* (Deckwer, 1992; Krishna, 1993); this regime is of importance in industrial reactor operation. With the aid of an extensive and dedicated experimental program of work, this paper aims to develop a unified description of the hydrodynamics of G–S fluid beds and G–L bubble columns, focusing mainly on the heterogeneous regime of operation. It is argued that appreciation of the *quantitative* hydrodynamic analogies can be a useful scale-up tool.

EXPERIMENTAL SET-UP AND SYSTEMS STUDIED

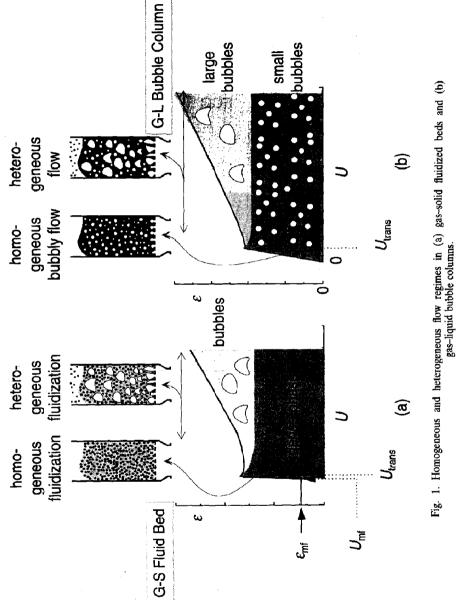
The experimental work was carried out in five columns made of polyacrylate sections. Two of these columns had a diameter of 0.1 m, one column had a diameter of 0.19 m and the remaining two columns were of 0.38 m diameter. All columns were equipped with sintered plate gas distributors. Figure 2 shows the layout of the 0.38 m column operating with FCC as the solid phase. This column was made of four sections each of 1 m length. Two cyclones, connected in series, were used to recover entrained fine particles and return these to the column. In G-L bubble column operation, the top section was open ended. In all cases, the pressure at the top of the column was close to atmospheric pressure. The gas inlet pipe at the bottom of the column was equipped with a quick shut-off valve for the purpose of performing dynamic gas disengagement experiments. A set of four rotameters was used for careful control and measurement of the gas flow rate to the column. Pressure taps were installed along the length of the column. Further details of the experimental set-up and the measurement techniques used can be found in Ellenberger (1994).

Table 1 gives a summary of the systems studied in the five columns.

TOTAL GAS HOLD-UP AND DYNAMIC GAS DISENGAGEMENT EXPERIMENTS

In G-S fluid bed operation the bed height was recorded visually. In G-L bubble column operation, high sensitivity pressure sensors were used to provide information on the gas hold-up, using the procedure discussed by Daly et al. (1992). For characterizing the heterogeneous flow regime in G-S and G-L systems, the dynamic gas disengagement experimental technique was used as discussed in the literature (Vermeer and Krishna, 1981; Daly et al., 1992; Shetty et al., 1992). The influence of disengagement of gas in the plenum chamber was confirmed to be negligibly small for all experiments (Grace, 1992). For the air-FCC system, a typical bed collapse experiment is shown in Fig. 3. The initial, quick disengagement of gas corresponds to the escape of the fast-rising bubbles in the fluid bed. This is followed by slow disengagement of the

Scale-up of fluidized bed and bubble column reactors





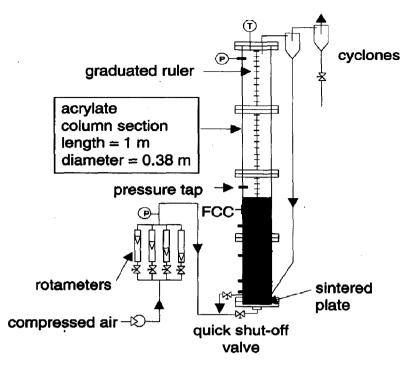


Fig. 2. Experimental set-up for a 0.38 m diameter G-S fluid bed.

Column diameter/ (m)	Distributor	Systems studied	Superficial velocity range (m/s)	Unexpanded bed height, H_0 (m)	Number of experiments
0.38	bronze ¹	air-FCC ³	0-0.382	0.1–1.5	238
0.19	bronze ¹	air-FCC ³	0-0.429	0.085-1.5	159
0.1	glass ²	air-FCC ³ He-FCC ³ Ar-FCC ³	00.196 00.385 00.166	0.2–1.1 0.55–1.1 0.21–1.1	200 199 202
0.38	bronze ¹	air–water ⁴ air–paraffin oil ⁵	0-0.71 0-0.066	0.5–2 0.9–1.5	460 239
0.19	bronze ¹	air–water ⁴ air–paraffin oil ⁵	0-0.65 0-0.64	0.37–1.1 0.7–1.5	209 293
0.1	glass ²	air-water ⁴ air-paraffin oil ⁵ air-tetradecane ⁶ He-tetradecane ⁶ Ar-tetradecane ⁶ SF ₆ -tetradecane ⁶	0-0.38 0-0.3 0-0.24 0-0.4 0-0.24 0-0.24	1.2 0.6–1.3 0.68–1.2 0.68–1.2 0.68–1.2 0.65–1.2	99 63 435 262 311 256

Table 1. Experimental set-up details, operating conditions and system properties

¹ Sintered bronze distributor with pore size = $50 \,\mu m$.

² Sintered glass distributor with pore size = $150-200 \ \mu m$.

³ Fluidized cracking caralyst: particle density, $\rho_p = 1,480 \text{ kg/m}^3$; bulk density, $\rho_{\text{bulk}} = 960 \text{ kg/m}^3$; cumulative particle size distribution: $10\% < 23 \mu \text{m}$; $50\% < 49 \mu \text{m}$; $90\% < 89 \mu \text{m}$. ⁴ Demineralized water: density, $\rho_L = 998 \text{ kg/m}^3$; dynamic viscosity, $\mu_L = 1 \text{ mPa s}$; surface tension,

 $\sigma = 72 \,\mathrm{mN/m}.$

⁵ Paraffin oil (special refined ISO paraffinic mineral oil with very low aromatic content): density, $\rho_L = 795 \text{ kg/m}^3$; dynamic viscosity, $\mu_L = 2.3 \text{ mPa}$ s; surface tension, $\sigma = 28 \text{ mN/m}$. ⁶ Tetradecane: density, $\rho_L = 763 \text{ kg/m}^3$; dynamic viscosity, $\mu_L = 2.2 \text{ mPa}$ s; surface tension,

 $\sigma = 27 \,\mathrm{mN/m}$.

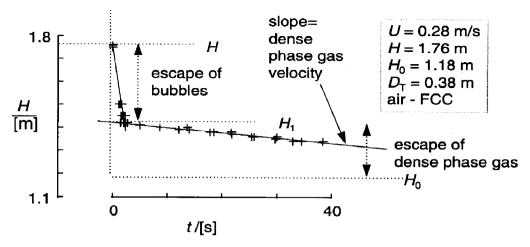


Fig. 3. Typical bed collapse experiments for gas-solid fluid beds with fluidized cracking catalyst as powder and air as fluidizing gas.

gas entrapped in the dense, or emulsion, phase. Due to the relatively slow disengagement characteristic of the dense phase gas, it was possible to record the bed height visually as a function of time.

From the graph shown in Fig. 3, the following parameters could be experimentally determined. The total gas voidage, or hold-up, ε , is

$$\varepsilon = 1 - \frac{\rho_{\text{bulk}}}{\rho_p} \frac{H_0}{H}, \qquad (1)$$

the hold-up of the bubbles, or "dilute" phase, ε_b , is determined from

$$\varepsilon_b = \frac{H - H_1}{H},\tag{2}$$

and the gas voidage in the "dense", or emulsion, phase is

$$\varepsilon_{df} = \frac{H_1 - H_0}{H_1} = \frac{\varepsilon - \varepsilon_b}{(1 - \varepsilon_b)}.$$
 (3)

The slope of the second, slowly disengaging, portion of the collapse curve was used to determine the superficial gas velocity through the dense phase, U_{df} .

For operation in the heterogeneous or churnturbulent regime of G-L bubble columns, typical disengagement experiments performed with water, tetradecane and paraffin oil are shown in Figs 4-6 respectively. In these G-L systems, the collapse experiments were monitored using the pressure sensors, following the procedure discussed by Daly *et al.* (1992). The total gas hold-up in gas-liquid systems was determined from

$$\varepsilon = 1 - \frac{H_0}{H} \tag{4}$$

The initial, fast-disengaging portions in Figs 4-6, were used to determine the hold-up of the "large" bubble population. Visual observation showed that the diameters of these large bubbles were of the order of 0.05-0.08 m. These "large" bubbles will subsequently be shown to be equivalent to "bubbles" or "dilute" phase in G-S fluid beds and, therefore, a common nomenclature is adopted here. The hold-up of the "large" bubbles is determined from eq. (2). Once the "large" bubbles have disengaged, the much smaller bubbles start disengaging. Typically the "small" bubbles are 2-5 mm in diameter and are strongly dependent on the physical properties of the system. For water and tetradecane, the validity of the assumption of a bimodal bubble size distribution, "large" and "small", is evident; see Figs 4 and 5. For the paraffinic mineral oil, also used in the experiments, severe foaming tendency was observed. "Small" and "micro" bubble disengagement regimes could be distinguished. The "micro" bubbles were typically smaller than about 1 mm in diameter. For the purposes of the analysis in this paper, which largely focuses on the "large" bubble, or "dilute" phase, the "small" and "micro" bubble populations are lumped together into a population which is termed the "dense" phase gas, in analogy with G-S fluid beds. Thus, the hold-up of the gas in the "dense" phase was determined using eq. (3). It is important to note here that the definition of "small" bubble hold-up in bubble columns in the literature (e.g. Deckwer and Schumpe, 1993) is different from the one used in eq. (3) for the "dense" phase gas voidage.

The slope of the disengagement curve for the "small" bubbles was used to determine the superficial gas velocity through the "dense" phase, U_{df} , neglecting the contribution of the "micro" bubbles. This neglect is justified because for paraffin oil, the

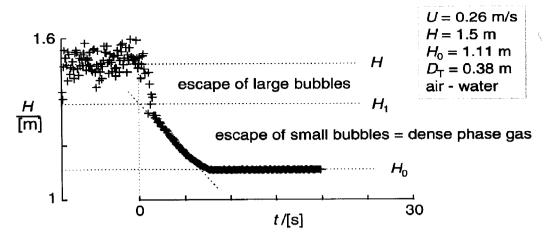


Fig. 4. Typical bed collapse experiments for gas-liquid bubble column with water as liquid phase and air as "fluidizing" medium.

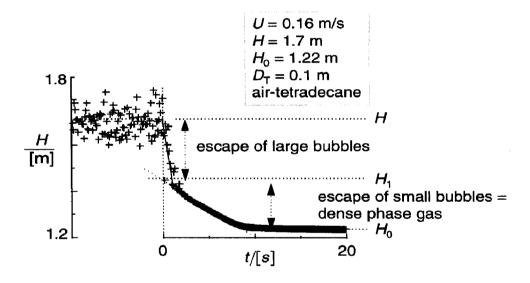


Fig. 5. Typical bed collapse experiments for gas-liquid bubble column with tetradecane as liquid phase and air as "fluidizing" medium.

superficial gas velocity through the "micro" bubbles is of the order of 0.005 m/s, much smaller than the superficial gas velocity through the "small" bubbles.

The analogous hydrodynamic picture used to analyze the experimental results is shown in Fig. 7, which presents the extension of the "two-phase" theory of G-S fluid beds to include G-L bubble columns. The superficial gas velocity through the dense phase $(U - U_{df})$ was determined by subtracting U_{df} from the superficial gas velocity U. In the experimental results reported below, the superficial gas velocity U was determined at the *average* pressure between the top and the bottom of the column. In total, about 4,000 total gas hold-up and dynamic gas disengagement experiments were performed with varying systems, superficial gas velocities, bed heights and column diameters.

TOTAL GAS VOIDAGE AND GAS VOIDAGE OF THE DENSE PHASE

For air-FCC, the dense phase gas voidage remains practically constant in the heterogeneous flow regime as shown in Fig. 8. This implies that a constant amount of gas, equal to U_{df} , is needed to keep the particles in suspension. The "excess" gas $(U - U_{df})$ rises up the column in the form of "dilute" phase or "bubbles". A typical plot of ε and ε_{df} versus the superficial gas velocity U,

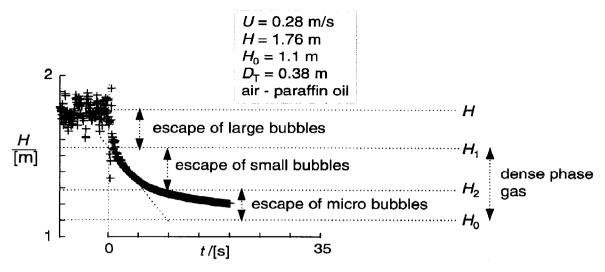


Fig. 6. Typical bed collapse experiments for gas-liquid bubble column with paraffin oil as liquid phase and air as "fluidizing" medium.

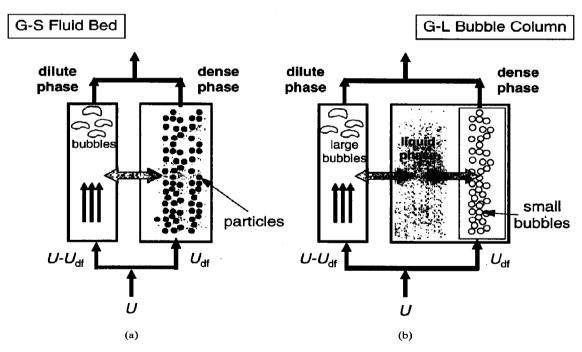


Fig. 7.(a) Classical two-phase model for gas-solid fluid beds after May (1959) and Van Deemter (1961). (b) Extension of two-phase model to bubble columns. The two-headed arrows represent the interphase mass transfer process.

obtained in the 0.38 m diameter column for air-FCC is shown in Fig. 8. Figures 9 and 10 show the corresponding plots for air-water and air-paraffin oil. For the gas-liquid systems the following analogy with air-FCC was noted: the voidage of the gas in the "dense" phase remains practically constant in the heterogeneous flow regime. At very high velocities, U > 0.5 m/s, the dense phase voidage tends to increase slightly. The behavior in the area of the transition region for air-FCC, air-water and air-paraffin oil (cf. Figs 8-10, respectively) is rather complex and was not investigated further in the current experimental study. The analysis to be further developed concentrates on the heterogeneous regime, clearly demarcated in Figs 8-10. The data of the 0.19 and 0.38 m diameter columns

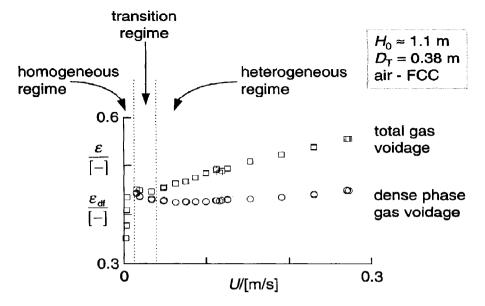


Fig. 8. Typical data for total gas voidage and gas voidage in the "dense" phase for a G-S fluid bed.

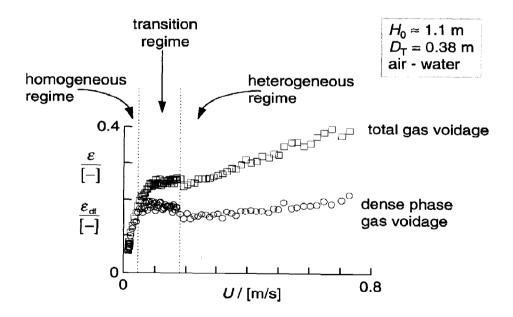


Fig. 9. Typical data for total gas voidage and gas voidage in the "dense" phase for a G-L bubble column with water as the liquid phase.

were fitted with identical sintered bronze distributors, and for these columns the dense phase gas voidage was independent of the column diameter, as witnessed in the results shown in Fig. 11. The swarm velocity of the dense phase gas, U_{ef}/ϵ_{df} , is also independent of the column diameter and practically constant in the heterogeneous flow regime; see Fig. 12. It should also be noted that the swarm velocity, U_{df}/ε_{df} , which equals the rise velocity of the "small" bubbles in G-L bubble columns, has a value of about 0.23 m/s for airwater, and a value of about 0.15 m/s for airparaffin oil.

On the basis of the insight generated in the

Scale-up of fluidized bed and bubble column reactors

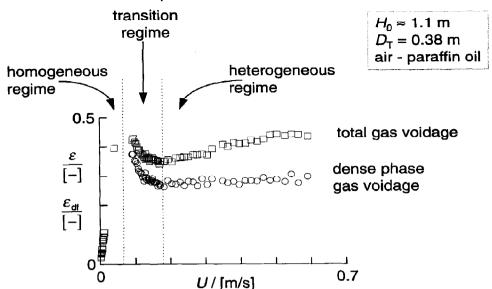


Fig. 10. Typical data for total gas voidage and gas voidage in the "dense" phase for a G-L bubble column with paraffin oil as the liquid phase.

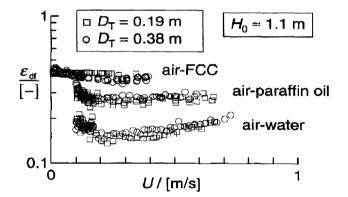


Fig 11. Voidage of dense phase gas ε_{df} versus the superficial gas velocity U for air-FCC, air-water air-water and air-paraffin oil. The results show that the swarm velocity is practically constant in the heterogeneous flow regime and is independent of the column diameter.

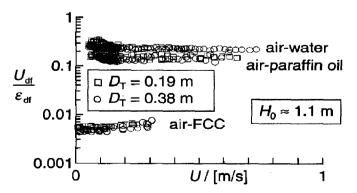


Fig. 12. Swarm velocity of dense phase gas (U_{df}/ϵ_{df}) versus the superficial gas velocity U for air-FCC, and air-paraffin oil. The results show that the dense phase gas voidage is practically constant in the heterogeneous flow regime and is independent of the column diameter.

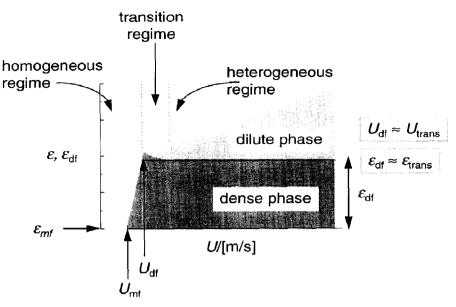


Fig. 13. Generalized model for gas hold-up in the heterogeneous flow regime.

preceding text a generalized model for the gas voidage in fluid beds and bubble columns can be set up; this model is shown schematically in Fig. 13. For estimation purposes, the gas voidage in the dense phase can be taken to be equal to the gas voidage at the regime transition point:

$$\varepsilon_{df} \approx \varepsilon_{\text{trans}}.$$
 (5)

The gas voidage at the regime transition point in gas-solid and gas-liquid systems can be predicted using a stability analysis (Batchelor, 1988; Biesheuvel and Gorissen, 1990; Krishna *et al.*, 1994). For gas-liquid bubble columns the correlations of Wilkinson *et al.* (1992) provide good estimates for the dense phase gas voidage

$$\varepsilon_{df} \approx \varepsilon_{\text{trans}} = \exp(-193\rho_G^{-0.61}\mu_L^{0.5}\sigma^{0.11}),$$
 (6)

and the dense phase superficial gas velocity $U_{df} \approx U_{\text{trans}}$

$$\frac{(U_{\text{trans}}/\varepsilon_{\text{trans}})\mu_L}{\sigma} = 2.25 \left(\frac{\sigma^3 \rho_L}{g\mu_L^4}\right)^{-0.273} \left(\frac{\rho_L}{\rho_G}\right)^{0.03}.$$
(7)

In order to predict the total gas hold-up, it remains to predict the hold-up of the "dilute" phase.

GAS HOLD-UP OF THE DILUTE PHASE

On the basis of the few thousand experiments carried out (see Table 1), it was noted that the dilute phase gas hold-up ε_b for *both* G–S and G–L systems is dependent on the total bed dispersion height H and on the column diameter D_T . For the

same values of $(U - U_{df})$ and H, a larger column diameter D_T results in lower ε_b . For the same values of $(U - U_{df})$ and D_T , a higher bed dispersion height leads to a lower value of ε_b . Though the influence of D_T and H on G-S fluid bed performance is well appreciated (e.g. Werther, 1983), the corresponding influence of these parameters on bubble column performance is not generally realized. For example, the correlation for the rise velocity of the "dilute" phase for G-S fluid beds developed by Worther (1983) shows a dependence $V_b \propto D_T^{0.4}$, while the corresponding correlation for the "large" bubble rise velocity in G-L bubble columns derived by Wilkinson et al. (1992) does not take account of the influence of column diameter. Before the experimental results for ε_b can be presented in a convenient and unified manner, it is necessary to develop a model which is capable of taking its dependence on H and D_T into account. The model that is developed here is derived by borrowing ideas and concepts which are well established for G-S fluid beds (Davidson et al., 1977; Van Swaaij, 1985) and relies on a proper description of bubble growth in the region above the distributor. This approach is used as a basis of the unified model for both G-S and G-L systems.

A model for dilute phase gas hold-up in G-S fluid beds

When a gas phase is injected into the grid of a G-S fluid bed, the jets formed initially at the distributor break up into bubbles; these bubbles then grow in size due to coalescence with the neighboring bubbles. For fast reactions such as combustion of coke on catalyst (as in the FCC regenerator), this initial bubble growth zone above

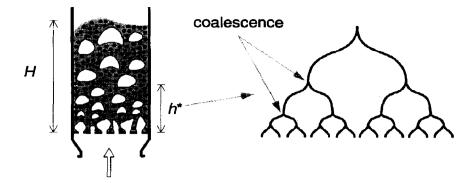


Fig. 14. Model for bubble growth in a gas-solid fluid bed (Darton et al., 1977).

the distributor is of great importance because of the very high mass transfer coefficients obtained. There is evidence in the literature to suggest that the bubbles do not grow indefinitely but reach a maximum "equilibrium" bubble size, d_b^* (Geldart, 1986). The growth of bubbles therefore terminates at a height h^* above the distributor where the equilibrium size d_b^* is attained; see Fig. 14 which is an adaptation of the model developed by Darton *et al.* (1977). The equilibrium bubble size d_b^* , and consequently the equilibration height h^* , are system properties, i.e. they depend on the powder used.

First, consider the zone $h = 0 - h^*$. Darton *et al.* (1977) suggested a bubble coalescence model depicted on the right of Fig. 14. In this model, coalescence occurs between bubbles of neighboring streams and the distance traveled by the bubbles before coalescence is proportional to their horizontal separation from neighboring bubbles. Following the Darton model, the diameter of a sphere having the same volume as the actual bubble is given by the relation:

$$d_b = \alpha_1 (U - U_{df})^{2/5} (h + h_0)^{4/5} g^{-1/5} \text{ for } h \le h^*. (8)$$

For dispersion heights exceeding h^* the bubble size remains constant and equals the equilibrium bubble size:

$$d_b^* = \alpha_1 (U - U_{df})^{2/5} (h^* + h_0)^{4/5} g^{-1/5} \text{ for } h^* \leq h \leq H$$
(9)

where α_1 is an empirical constant; this constant was determined by Darton *et al.* (1977) by fitting measured experimental data and they found its value to be 0.54 for FCC powder. The parameter h_0 characterizes the distributor; the initial bubble size formed at the distributor is thus:

$$d_{b0} = \alpha_1 (U - U_{df})^{2/5} (h_0)^{4/5} g^{-1/5}.$$
(10)

The value of h_0 can be estimated from

$$h_0 = 4\sqrt{A_0} \tag{11}$$

where A_0 represents the "catchment area" of the bubble stream at the distributor plate and is usually the area of the plate per orifice (nozzle). For porous plate distributors, for example, $A_0 =$ 0.000056 m², giving $h_0 = 0.03$ m; this value is used in all the model calculations and correlations to be presented in this paper.

The hold-up of gas in the form of bubbles, i.e. the dilute phase of a bubbling bed of height H, can be calculated from the following relation

$$\varepsilon_b = \frac{1}{H} \int_0^H \frac{(U - U_{df})}{V_b} dh \qquad (12)$$

where the rise velocity of the bubbles is a function of the bubble diameter, d_b . Werther (1983) carried out extensive measurements in beds of different diameters and found that the rise velocity of a bubble of a given size was dependent on the size of the vessel D_T ; a bubble tends to rise faster in a larger diameter bed. The increase of V_b with D_T is due to the large-scale mixing patterns that have the effect of accelerating the bubble swarms. These large-scale mixing patterns are caused by eddies whose size is limited by the vessel diameter. Werther (1983) proposed the following correlation for the bubble rise velocity:

$$V_b = \phi \sqrt{gd_b} \tag{13}$$

where ϕ is an empirical constant that depends on the column diameter. For A type powders, such as fluidized cracking catalyst, ϕ was correlated by Werther as:

$$\begin{aligned}
\phi &= 1.0 & \text{for } D_T \leq 0.1 \text{ m} \\
\phi &= 2.5 D_T^{0.4} & \text{for } 0.1 \leq D_T \leq 1 \text{ m} \\
\phi &= 2.5 & \text{for } D_T \geq 1 \text{ m}.
\end{aligned}$$
(14)

For B type powders, such as sand, the Werther correlation is:

$$\phi = 0.64 for D_T \le 0.1 m
\phi = 1.6D_T^{0.4} for 0.1 \le D_T \le 1 m (15)
\phi = 1.6 for D_T \ge 1 m.$$

For analysis and correlation of the experimental data generated in this work, in columns of diameter 0.1, 0.19 and 0.38 m, the following form was adopted for the rise velocity constant ϕ :

$$\phi = \phi_0 D_T^n \tag{16}$$

where the constant ϕ_0 and the exponent *n* are experimentally determined parameters.

Combining eqs (8), (9), (12), (13) and (16) the following expressions can be derived for the dilute phase gas hold-up for, respectively, dispersion heights H smaller and greater than the equilibration height h^* :

yielded $h^* = 1.16(U - U_{df})^{0.33}$ which was only marginally worse than the linear fit reported above. Figure 15 compares the obtained fit for the rise velocity constant $\phi = 1.95D_T^{1/3}$ with the Werther correlation (1983); the correspondence is remarkably close. The fitted dependence of the equilibration height H^* on $(U - U_{df})$ is portrayed in Fig. 16. The values of h^* range between 0.4 and 1.2 m; this is within the range of h^* values assumed by Werther (1983) to rationalize the observed mass transfer data for a fluid bed reactor using silica catalyst. Since the value of the equilibration height h^* increases with increasing values of $(U - U_{df})$, we

$$\varepsilon_b = \frac{1}{\sqrt{\alpha_1} \phi_0 D_T^n g^{2/5}} \frac{\left[(H+h_0)^{3/5} - (h_0)^{3/5} \right]}{(3/5)} \frac{(U-U_{df})^{4/5}}{H} \text{ for } H \le h^*$$
(17a)

and

$$\varepsilon_{b} = \frac{1}{\sqrt{\alpha_{1}} \phi_{0} D_{T}^{n} g^{2/5}} \frac{\left[(h^{*} + h_{0})^{3/5} - (h_{0})^{3/5} \right] (U - U_{df})^{4/5}}{(3/5)} H + \frac{1}{\sqrt{\alpha_{1}} \phi_{0} D_{T}^{n} g^{2/5}} (h^{*} + h_{0})^{-2/5} (H - h^{*}) \frac{(U - U_{df})^{4/5}}{H} \text{ for } H > h^{*}.$$
(17b)

The model parameters in eq. (17), h^* , α_1 , ϕ_0 and *n* can be determined by fitting with experimental data.

Correlation of dilute phase hold-up data for air-FCC fluid bed

There is no a priori reason to assume that the equilibration height h^* would be independent of the superficial gas velocity through the dilute phase $(U-U_{df})$; therefore, the experimental data were fitted by assuming three different forms for the dependence of h^* on $(U - U_{df})$: (1) a linear dependence: $h^* = b_0 + b_1(U - U_{df})$, (2) an exponential dependence: $h^* = b_0 \exp[b_1(U - U_{df})]$, and (3) a power-law dependence: $h^* = b_0 (U - U_{df})^{b_1}$. The linear and exponential fits for h^* were equally accurate and slightly superior to the power law. Furthermore, the constant in the Darton bubble growth model was fixed at the value $\alpha_1 = 0.54$ for the FCC powder used in the experiments. The few hundred experimental data for ε_b obtained for air-FCC in 0.1, 0.19 and 0.38 m diameter columns with varying values for $(U - U_{df})$ and H were used to find the best fit values for the model parameters ϕ_0 , b, b_0 and b_1 using a non-linear regression routine. The results of the regression analysis yield the following values for the various model parameters in eq. (17):

$$\alpha_1 = 0.54; h^* = 0.33 + 1.51(U - U_{df});$$

 $\phi = 1.95D_T^{1/3}$ (air-FCC). (18)

The power-law fit for the equilibrium height

should expect the equilibration bubble size d_b^* to also increase with increasing $(U - U_{df})$. Visual observations in a two-dimensional column confirmed this trend.

The importance of taking bubble growth into account is demonstrated by the calculations presented in Fig. 17, for $(U - U_{df}) = 0.3$ m/s and three column diameters $D_T = 0.1$, 0.5 and 1 m. The dilute phase hold-up ε_b decreases with increasing bed dispersion H; this decrease is particularly significant in the range 0.5–2 m. Beyond about H = 4 m there is practically no further influence on bed dispersion height. For $H \gg h^*$, eq. (17b) yields the asymptotic value for the dilute phase hold-up, ε_b :

$$\varepsilon_b = \frac{1}{\sqrt{\alpha_1} \phi_0 D_T^n g^{2/5}} (h^* + h_0)^{-2/5} (U - U_{df})^{4/5}$$
for $H \gg h^*$. (17c)

The dilute phase hold-up also decreases with increasing column diameter; cf. Fig. 17. According to the Werther correlations, eqs (14) and (15), the column diameter dependence vanishes after $D_T = 1$ m; this aspect has not yet been verified experimentally in the literature on G-S fluid beds.

In order to demonstrate the validity of the model represented by eq. (17), and to test the goodness of the fitted parameters given by eq. (18) a small selection of the experimental data will be presented in order to show the influence of the dispersion height H on the hold-up of the dilute phase. In Fig. Scale-up of fluidized bed and bubble column reactors

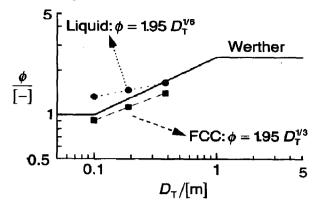


Fig. 15. The dependence of the rise velocity constant ϕ on the column diameter. Correlations for air-FCC and air-liquids compared with the Werther correlation of eqs (14) and (15).

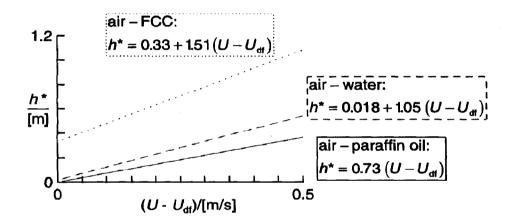


Fig. 16. The influence of $(U - U_{df})$ on the height at which the (large) bubbles reach their equilibrium size. Fitted correlations for air-FCC, air-paraffin oil and air-water.

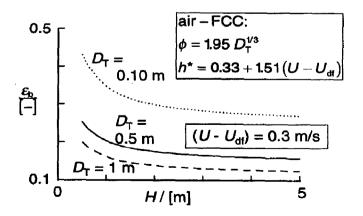


Fig. 17. Simulations of dilute phase hold-up for air-FCC using the model eq. (17). The results show strong dependence on the bed dispersion height and on the column diameter.

J. ELLENBERGER and R. KRISHNA

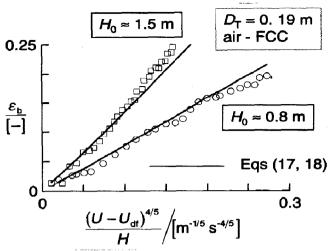


Fig. 18. The "dilute" phase hold-up in air-FCC fluid beds in a 0.19 m diameter column and for two different values of the unexpanded bed, H_0 .

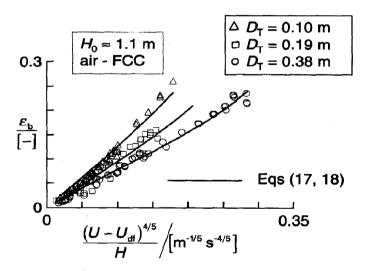


Fig. 19. The "dilute" phase hold-up in air-FCC fluid beds showing dependence on the column diameter.

18, data obtained in the 0.19 m column for unexpanded bed heights of $H_0 = 0.8$ and 1.5 m are compared. The plot of ε_b versus $(U - U_{df})^{4/5}/H$ is motivated by the form of the expression (17). The fitted curve represents the data very well. The observed trend in each individual data series was found to be well represented by eq. (17). When compared at the same value of $(U - U_{df})$, the dilute phase hold-up ε_b decreases with increasing dispersion height; cf. Fig. 17.

The influence of column diameter on the dilute phase hold-up is portrayed in Fig. 19. Here, data for the 0.1, 0.19 and 0.38 m diameter columns obtained with the same ungassed bed height $H_0 = 1.1$ m are compared. The experimental results show the significant influence of the column diameter. Figure 19 also verifies the validity of the experimental fit [eq. (18)].

Correlation of dilute phase hold-up data for air-paraffin oil bubble column

Taking encouragement from the observed, albeit superficial, similarities in the total gas voidage and dense phase voidages obtained in G-S fluid beds and G-L bubble columns (cf. Figs 8-10), it will be assumed that the dilute phase gas hold-up (= large bubble population), ε_b , in the bubble column with paraffin oil as the liquid phase, also conforms to the model represented by Fig.14 and quantitatively by eq. (17). Essentially, this implies that large bubbles in G-L systems are formed by coalescence of small bubbles and that this coalescence is restricted to

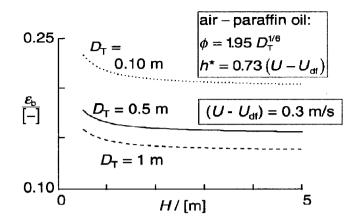


Fig. 20. Simulations of dilute phase hold-up for air-paraffin oil using the model eq. (17). The results show dependence on the bed dispersion height and on the column diameter.

within the equilibration height h^* above the distributor. This assumption will be confronted with experimental data later to test its validity. The analysis of Jamialahmadi and Müller-Steinhagen (1993) shows the applicability of eq. (13) to describe the rise velocity of the "large" bubbles. Following an identical procedure, the several hundred experimental data points were subjected to a non-linear regression analysis to obtain values of the model parameters h^* , α_1 , ϕ_0 and n. An additional complication arises for G-L systems because, in contrast to the situation for FCC for which the bubble growth parameter value $\alpha_1 = 0.54$, the value of α_1 is unknown. It is therefore only possible to fit the parameter $\sqrt{\alpha_1} \phi$. As with air-FCC, a linear dependence of h^* on $(U-U_{df})$ was found to yield the best results. Arbitrarily assuming that $\alpha_1 = 1$, the model parameters were found to be:

$$\alpha_1 = 1, h^* = 0.73(U - U_{df}); \phi = 1.95 D_T^{1/6}$$

(air-paraffin oil). (19)

In Fig. 15, the rise velocity constant ϕ for air-paraffin oil is compared with that found earlier for air-FCC; it can be observed that the diameter dependence of the rise velocity of the dilute phase, V_b is weaker for paraffin oil. In Fig. 16, the equilibration height obtained for air-paraffin oil is compared to that obtained for air-FCC and it is noted that the equilibration height for air-paraffin oil is significantly lower. Therefore, the dilute phase hold-up ε_b should be expected to show a weaker dependence on the dispersion height than for FCC. This is verified by the calculations shown in Fig. 20. It is clear that influence of the dispersion height on the bubble hold-up tends to disappear only beyond H = 3 m. Comparison of the analogous Figs 17 and 20 provides justification for the title of this paper; it is indeed possible to approach the scale-up of G-S fluid bcds and G-L bubble columns using a unified approach based on the physical model represented by Figs 7 and 14.

Figures 21 and 22 present the quantitative justification of the model represented by Fig. 14 and eq. (17), and underline the analogies with the air-FCC; cf. Figs 18 and 19.

Correlation of dilute phase hold-up data for air-water bubble column

The procedure followed for air-paraffin oil can be repeated for air-water. The model parameters are found to be:

$$\alpha_1 = 1; h^* = 0.018 + 1.05(U - U_{df});$$

 $\phi = 1.95 D_T^{1/6}$ (air-water). (20)

The rise velocity constant ϕ and the equilibration heights h^* are compared with the corresponding parameters obtained for air-FCC and air-paraffin oil in Figs 15 and 16. The only difference between air-water and air-paraffin oil appears to be the slightly higher value for the equilibration height h^* ; otherwise the conclusions for the column diameter and dispersion height dependencies are similar to that portrayed in Fig. 20.

Figures 23 and 24 provided quantitative verification of the goodness of fit of the model [eq. (17)] to the measured experimental data.

Influence of gas density on bed voidage and dilute phase hold-up

For G–S fluid bed of 0.1 m diameter with FCC as powder, measurements were carried out with helium, air and argon as fluidizing gas. Figure 25 compares the results obtained with these three gases on the total gas voidage. The total gas voidage increases with increasing gas density. The influence of increased gas density is to delay transition to the heterogeneous regime (Krishna *et*

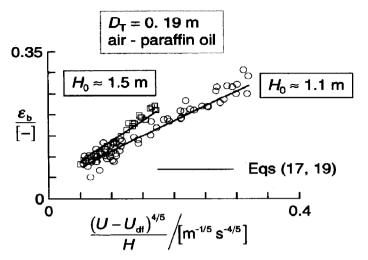


Fig. 21. The "dilute" phase hold-up in air-paraffin oil bubble beds in a 0.19 m diameter column and for two different values of the unexpanded bed, H_0 .

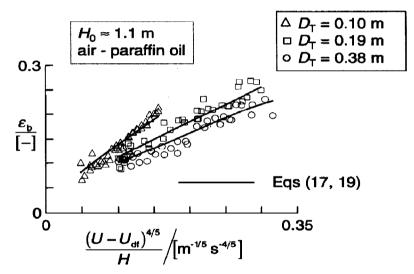


Fig. 22. The "dilute" phase hold-up in air-paraffin oil bubble column showing dependence on column diameter.

al., 1993), i.e. increased ρ_G tends to increase $\varepsilon_{\text{trans}}$ ($\approx \varepsilon_{df}$) and the increase in the total gas voidage is to be attributed to increase in the gas voidage of the dense phase. By implication, the influence of gas density on the dilute phase hold-up ε_b would be negligibly small; this expectation is confirmed in Fig. 26. The continuous curve in Fig. 26 represents the model prediction of eq. (17) with the parameters given in eq. (18). The convenient result portrayed in Fig. 26 suggests that experiments carried out at ambient pressure conditions could provide a good basis for predicting the dilute phase hold-up at high pressure.

In the 0.1 m diameter bubble column, experiments were carried out with tetradecane as the liquid phase and with helium, air, argon and sulfur hexafluoride (SF₆) as the gas phase. The results for the total gas voidage shows a pronounced increase of the total gas voidage with increasing gas density, in analogy with results obtained for FCC; compare the results of Fig. 27 with those of Fig. 25. The dilute phase hold-up ε_{b} , on the other hand, shows no particular trend with the gas density; see Fig. 28 which is endogenous with Fig. 27. The rationalization of the results in Fig. 27 and Fig. 28 is that increased gas density tends to delay transition to the heterogeneous flow regime and thus increase the value of the dense phase gas voidage ε_{df} ($\approx \varepsilon_{trans}$); the dilute phase gas hold-up is unaffected. This conclusion was reached earlier by

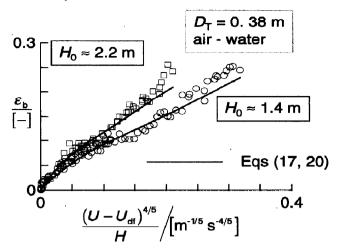


Fig. 23. The "dilute" phase hold-up in air-water bubble columns in a 0.19 m diameter column and for two different values of the unexpanded bed, H_0 .

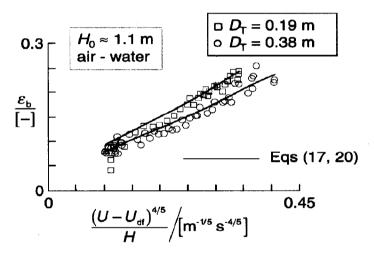


Fig. 24. The "dilute" phase hold-up in air-water bubble columns showing dependence on column diameter.

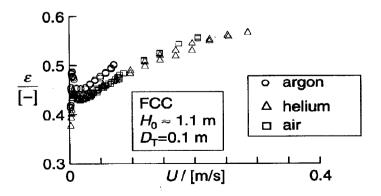


Fig. 25. Influence of varying gas density on the total gas voidage in a G-S fluid bed of 0.1 m diameter with FCC as powder.

5407

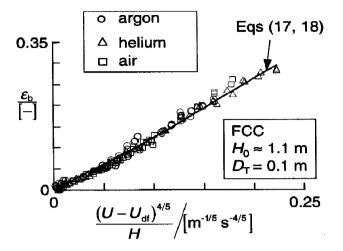


Fig. 26. Demonstration of the independence of the dilute phase hold-up on the gas density in a 0.1 m diameter fluid bed operation with FCC as powder.

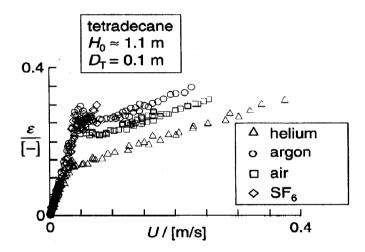


Fig. 27. Influence of varying gas density on the total gas voidage in a 0.1 m diameter G-L bubble column with tetradecane as the liquid phase.

Krishna et al. (1991) from an analysis of the data of Wilkinson (1991). The two continuous curves shown in Fig. 28 are drawn using the fitted parameters for the air-paraffin oil and air-water systems. The differences in the predictions are small, suggesting that the liquid properties do not significantly influence the dilute phase hold-up.

Influence of liquid properties on the dilute phase gas hold-up in bubble columns

From Figs 9 and 10 it is observed that the total gas voidage for the water and paraffin oil systems are significantly different. However, if the dilute phase hold-ups for these two systems are compared, it can be seen that this parameter varies only slightly; see Fig. 29. That the influence of liquid phase properties has only a minor influence on the dilute phase hold-up is confirmed by examination of literature data published by Grund (1988) and Krishna *et al.* (1991) for the water, methanol, ligroin, toluene, turpentine, butanol and glycol systems; see Fig. 30. Despite a wide variation in viscosity and surface tension, the dilute phase hold-up ε_b is only slightly affected. Figure 30 also shows the calculations using eqs (17) and (19), and eqs (17) and (20), derived for paraffin oil and water, respectively, and confirms the ability of the model developed in this paper to correlate literature data not used in the fitting of the model parameters.

CONCLUSIONS

The results presented in this paper support the authors' contention that a unified approach to the

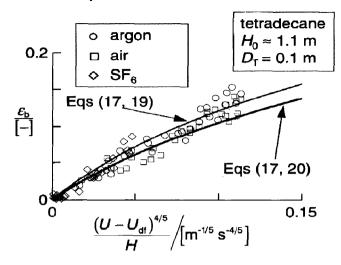


Fig. 28. Demonstration of the independence of the dilute phase hold-up on the gas density in a 0.1 m diameter bubble column with tetradecane as the liquid phase. The two continuous curves are drawn using eqs (17) and (19), and eqs (17) and (20).

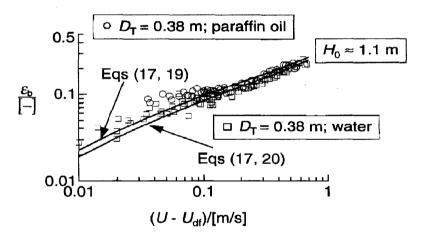


Fig. 29. Comparison of the dilute phase hold-up for paraffin oil and water in a 0.38 m diameter column.

scale-up of G–S fluid beds and G–L bubble columns is possible. This unified approach is based on hydrodynamic analogies in the behavior of these two systems. On the basis of extensive data generated for G–S fluid beds and G–L bubble columns, the following analogies between the hydrodynamic behavior of these two columns have been established.

- The hydrodynamic behavior of fluid beds and bubble columns can be modeled using the generalization of the two-phase model developed for fluid beds; see Fig. 7.
- (2) For both fluid beds and bubble columns, the dense phase gas voidage ε_{df} is practically

independent of the column diameter. This parameter shows an increasing trend with increasing gas density.

- (3) The dilute phase hold-up in both fluid beds and bubble columns can be described by eq.
 (17) which is derived from a coalescence model put forward by Darton *et al.* (1977) for gas-solid fluid beds.
- (4) The rise velocity of the dilute phase V_b for both fluid beds and bubble columns increases with column diameter [cf. eq. (16)]; this dependence is observed to be stronger for air-FCC than for air-paraffin oil, air-water and air-tetradecane.
- (5) The dilute phase hold-up for G-S and G-L

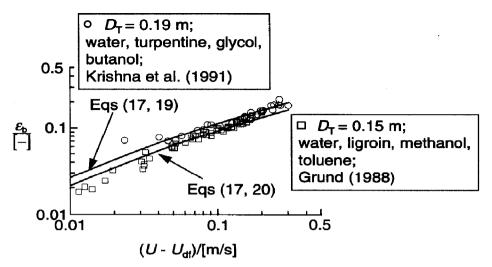


Fig. 30. Influence of liquid properties on the dilute phase hold-up.

systems tends to decrease with increasing dispersion heights—a consequence of bubble coalescence. The equilibration height h^* is found to be an increasing function of the superficial gas velocity through the dilute phase, $(U - U_{df})$. The values of h^* for air–FCC are significantly higher than for air–paraffin oil and air–water. As a consequence, the influence of the dispersion height H on the dilute phase hold-up is stronger for air–FCC than for air–liquids; compare the results of Figs 17 and 20.

(6) The difference in the dilute phase hold-up with liquid properties is very small. The values of ε_b obtained with tetradecane agree with that obtained with paraffin oil, even though these two systems show widely different total gas voidages; see Figs 9, 10 and 29. As an approximation, it may be assumed that the influence of gas density and liquid properties on the hydrodynamics of G-L bubble columns is only on the dense phase voidage ε_{df} and the dense phase superficial gas velocity U_{df} .

The extension of the unified approach to the modeling of the hydrodynamics of G-S fluid beds and G-L bubble columns to include interphase mass transfer is the next research item in the authors' comprehensive agenda.

Acknowledgements—The infrastructural facilities for the experimental work were created with a general financial grant from the University of Amsterdam. The authors acknowledge the support received from Prof. P. W. M. de Meijer and Prof. A. Bliek in this regard. The experimental facilities were built with the assistance of J. Zoutberg, D. P. de Zwarte, Th. J. A. M. Nass and W. H. Buster of the mechanical workshop of the Chemical Engineering Department. G. C. N. van den Aardweg

provided valuable organizational assistance. The experimental work reported in this paper was performed with the assistance of M. Benrahmoun, A. de Draaijer, L. Fan, R. Floore, W. L. Ghijsen, M. van der Gun, D. E. Hennephof, P. Kouwenhoven and A. Roest.

NOTATION

A_0	catchment area of single orifice, m ²
b_0	fir parameter for equilibration height, m
b_1	fit parameter for equilibration height
d_b	bubble diameter of dilute phase, m
d_{b0}	initial bubble size formed at distributor,
<i>ub</i> 0	m
d_b^*	equilibrium bubble size of dilute phase,
цb	m
د	
d_p	mean particle size, m
D_{ax}	axial dispersion coefficient, m ² /s
D_T	column diameter, m
g	acceleration due to gravity, 9.81 m/s ²
g h	height above the gas distributor, m
h*	height above the gas distributor where
	the bubbles reach their equilibrium size,
	m
h_0	parameter determining the initial bubble
()	size at the gas distributor, m
н	height of expanded bed, m
H_0	height of unexpanded bed, m
H_1	height of dispersion after escape of di-
	lute phase, m
n	power in the rise velocity correlation eq.
	(16)
U	superficial gas velocity, m/s
$(U-U_{df})$	superficial gas velocity through the di-
	lute phase, m/s
U_{df}	superficial velocity of gas thrugh the
-	dense phase, m/s

 U_{mf} minimum fluidization velocity for gassolid fluid bed, m/s

U_{mb}	minimum "bubbling" velocity for	gas-
	solid fluid bed, m/s	
11	cuperficial and velocity at regime t	ronei

- Utrans superficial gas velocity at regime transition, m/s
- V_b rise velocity of the dilute phase, m/s

Greek letters

constant in the bubble growth model,
eq. (8)
total gas voidage of G-S or G-L system
gas hold-up in "dilute" phase
gas hold-up in "dense" phase
voidage of G-S fluidized bed at mini-
mum fluidization conditions
gas hold-up at the regime transition
point
gas viscosity, Pa s
liquid viscosity, Pa s
bulk density of particles, kg/m ³
density of gaseous phase, kg/m ³
liquid density, kg/m ³
particle density, kg/m ³
surface tension of liquid phase, N/m
rise velocity constant in eq. (13)
constant defined in eq. (16)

Subscripts

b	referring to "dilute" phase
df	referring to "dense" phase
G	referring to gas phase
L	referring to liquid phase
mf	referring to minimum fluidization condi-
	tions
р	referring to particle phase
trans	referring to the transition point
0	referring to conditions at the gas distri-
	butor $(h = 0)$

Superscripts

referring to equilibrium value

REFERENCES

- Batchelor, G. K., 1988, A new theory of the instability of a uniform fluidized bed. J. Fluid. Mech. 193, 75–110.
- Biesheuvel, A. and Gorissen, W. C. M., 1990, Void fraction disturbances in a uniform bubbly fluid. Int. J. Multiphase Flow 16, 211–231.
- Daly, J. G., Patel, S. A. and Bukur, D. B., 1992, Measurement of gas holdups and sauter mean diameters in bubble column reactors by dynamic gas disengagement method. *Chem. Engng Sci.* 47, 3647– 3654.
- Darton, R. C., LaNauze, R. D., Davidson, J. F. and Harrison, D., 1977, Bubble growth due to coalescence in fluidized beds. *Trans. Inst. Chem. Engrs* 55, 274–280.
- Davidson, J. F., Harrison, D., Darton, R. C. and LaNauze, R. D., 1977, The two-phase theory of fluidization and its application to chemical reactors, in *Chemical Reactor Theory, A Review* (Edited by L. Lapidus and N. R. Amundson), pp. 583-685. Prentice-Hall, Englewood Cliffs, New Jersey.
- Deckwer, W.-D., 1992, Bubble Column Reactors. John Wiley, New York.
- Deckwer, W.-D. and Schumpe, A., 1993, Improved tools for bubble column reactor design and scale-up. Chem.

Engng Sci. 48, 889-911.

- Ellenberger, J., 1994, Experimental set-up for studies in multiphase reactor hydrodynamics, Internal Report, Department of Chemical Engineering, University of Amsterdam, Amsterdam, The Netherlands.
- Geldart, D., (ed.), 1986, Gas Fluidization Technology. John Wiley, New York.
- Grace, J. R., 1992, Agricola aground: characterization and interpretation of fluidization phenomena. A.I.Ch.E. Symposium Series 88(289), 1-16.
- Grund, G., 1988, Hydrodynamische Parameter und Stoffaustauscheigenschaften in Blasensäulen mit organischen Medien. Ph.D. dissertation, University of Oldenburg, Germany.
- Grund, G., Schumpe, A. and Deckwer, W.-D., 1992, Gas-liquid mass transfer in a bubble column with organic liquids. *Chem. Engng Sci.* 47, 3509-3516.
- Jamialahmadi, M. and Müller-Steinhagen, H., 1993, Gas hold-up in bubble column reactors, in *Encyclopedia of Fluid Mechanics*, Supplement 2, Advances in Multiphase Flow (Edited by N. P. Cheremisinoff), pp. 387-407. Gulf Publishing, Houston.
- Krishna, R., 1993, Analogies in multiphase reactor hydrodynamics, in *Encyclopedia of Fluid Mechanics*, Supplement 2, Advances in Multiphase Flow (Edited by N. P. Cheremisinoff), pp. 239–297. Gulf Publishing, Houston.
- Krishna, R., Wilkinson, P. M. and Van Dierendonck, L. L., 1991, A model for gas hold-up in bubble columns incorporating the influence of gas density on flow regime transitions. *Chem. Engng Sci.* 46, 2491–2496.
- Krishna, R., Ellenberger, J. and Hennephof, D. E., 1993, Analogous description of gas-solid fluidized beds and bubble columns. *Chem. Engng. J.* 53, 89-101.
 Krishna, R., de Swart, J. W. A., Hennephof, D. E.,
- Krishna, R., de Swart, J. W. A., Hennephof, D. E., Ellenberger, J. and Hoefsloot, H. C. J., 1994, Influence of increased gas density on hydrodynamics of bubble column reactors. A.I.Ch.E. J. 40, 112–119.
- May, W. G., 1959, Fluidized bed reactor studies. Chem. Engng Prog. 55, 49-56.
- Miyauchi, T., Furusaki, S., Morooka, S. and Ikada, Y., 1981, Transport phenomena and reaction in fluidized catalyst beds, in *Advances in Chemical Engineering*, Vol. II (Edited by T. B. Drew, G. R. Cokelet, J. W. Hoopes, Jr. and T. Vermeulen), pp. 275-448. Academic Press, New York.
- Schumpe, A. and Grund, G., 1986, The gas disengagement technique for studying gas holdup structure in bubble columns. *Canad. J. Chem. Engng* 64, 891-896.
- Shetty, S. A., Kantak, M. V. and Kelkar, M. G., 1992, Gas-phase backmixing in bubble column reactors. *A.I.Ch.E. J.* 38, 1013–1026.
- Van Deemter, J. J., 1961, Mixing and contacting in gas-solid fluidized beds. Chem. Engng Sci. 13, 143-154.
- Van Swaaij, W. P. M., 1985, Chemical reactors, in *Fluidization*, 2nd edn (Edited by J. F. Davidson, R. Clift and D. F. Harrison), pp. 595-629. Academic Press, New York.
- Vermeer, D. J. and Krishna, R., 1981, Hydrodynamics and mass transfer in bubble columns operating in the churn-turbulent regime. Ind. Engng Chem. Process Design & Dev. 20, 475-482.
- Werther, J., 1983, Hydrodynamics and mass transfer between bubble and emulsion phase in fluidized beds of sand and cracking catalyst, in *Fluidization, IV* (Edited by D. Kunii and R. Toei), pp. 93–102. Engineering Foundation, New York.
- Wilkinson, P. M., 1991, Physical aspects and scale-up of high pressure bubble columns. Ph.D. dissertation (Chemical Engineering), (University of Groningen, Groningen, The Netherlands.
- Wilkinson, P. M., Spek, A. P. and Van Dierendonck, L. L., 1992, Design parameters estimation for scale-up of high-pressure bubble columns. A.I.Ch.E. J. 38, 544-554.