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# REACTOR DEVELOPMENT FOR CONVERSION OF NATURAL GAS TO LIQUID FUELS: A SCALE-UP STRATEGY RELYING ON HYDRODYNAMIC ANALOGIES

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**Abstract** - Gas-solid fluidized beds and bubble column slurry reactors are very commonly encountered in processes for conversion of natural gas to liquid fuels and light olefins. This paper proposes a unified design and scale-up strategy for such "fluidized" multiphase reactors. The approaches uses hydrodynamic analogies as a basis. These analogies have been investigated in a variety of gas-solid and gas-liquid systems in several columns varying in diameters between 0.05 m and 0.63 m. It is argued that the appreciation of the hydrodynamic analogies will allow cross-fertilisation of concept and design data, thereby reducing process development costs.

## INTRODUCTION

Fluidized bed and bubble column reactors are finding increasing use in industrial practice; these two reactor technologies figure prominently in processes for converting natural gas to liquid fuels and light olefins; cf. Fig. 1. There are considerable reactor design and scale-up problems associated with these natural gas conversion technologies; these problems arise because of several special features of these processes. Firstly, large gas throughputs are involved, necessitating the use of large diameter reactors, typically 5-8 m, often in parallel. Secondly, the processes operate under high pressure conditions, typically 10-80 bar. Thirdly, in order to obtain high conversion levels, large reactor heights, typically 20-30 m tall, are required. Finally, most of these processes are exothermic in nature, requiring heat removal by means of cooling tubes inserted in the reactor. Successful commercialisation of natural gas conversion technologies are crucially dependent on the proper understanding of the scaling up principles of fluid beds and bubble columns for these conditions which fall outside the purview of most published theory and correlations.

A commonly used scale-up strategy for fluidized beds and bubble column reactors makes a separation between the more chemistry oriented "process research" and the more physics/mechanics oriented "engineering research". Thus conversion of actual feedstocks with real catalysts under representative conditions of temperature, pressure and space velocity are studied in relatively small reactor systems (e.g. bench scale units) that are considerably scaled down versions of the (envisaged) commercial plant while hydrodynamics, mass and heat transfer, residence time distributions, attrition of catalyst particles etc. are studied on a representative, not necessarily small scale in so-called cold models, or "mock ups", that are

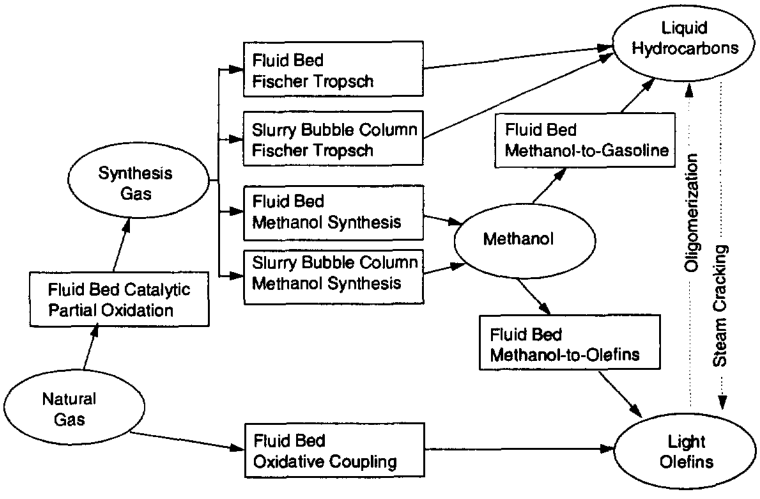

Fig. 1. Fluid bed and bubble column reactor technologies in processes for converting natural gas to liquid fuels and light olefins.

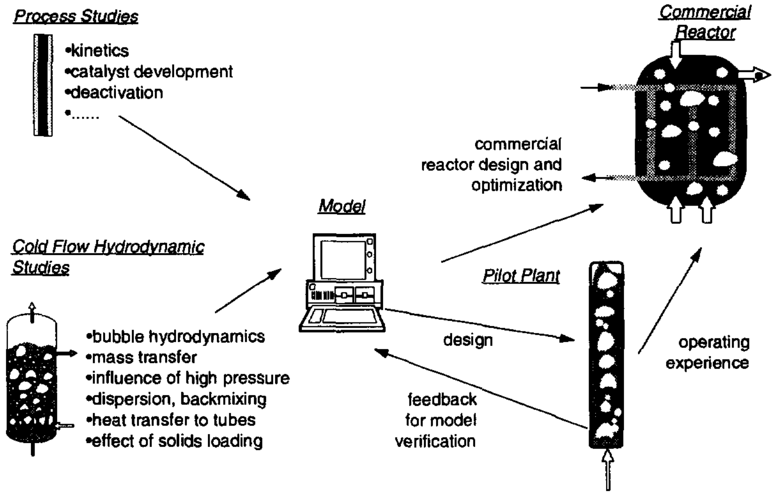

Fig. 2. Scale up strategy for fluid bed and bubble column reactors.

more easily available and more convenient to use (Euzen *et al.*, 1993). These cold models, together with the hot process units and computational models are the main tools in reactor scale-up. Figure 2 sketches the commonly used scale-up strategy for commercialisation of fluid beds and bubble columns and shows how the information generated in the process and engineering research studies can be integrated by means of mathematical models. These models allow extrapolations outside the area where the experimental data can be generated and thus they can be used to link the engineering information obtained in cold flow models that are reasonably scale representative but different in temperature, pressure and nature of the materials, with the process data from reactor data system that are representative in the latter respects but which deviate considerably in scale. This way of integrating can in many cases form a sufficient basis for a definition of the desired process. Further confirmation of the integrated operation of the process can, if deemed necessary, be obtained in studies using a larger and/or more complex pilot plant, which can be designed on the basis of the information obtained in the above studies. Depending upon whether flexibility in varying process parameters or true simulation of the integrated process is considered more important, this pilot plant can be either a relatively small, flexible pilot plant or a near-commercial size demonstration plant which is generally designed on the basis of a rather narrow process specification.

In this paper we focus on "cold flow" engineering research and suggest a strategy for cutting development time and costs by appreciation of the analogies in the hydrodynamics, mass transfer and mixing characteristics of fluid beds and bubble columns. In order to demonstrate these analogies we undertook a comprehensive experimental program of investigation of these two reactor types.

## EXPERIMENTAL

### *Gas-solid fluidized bed experiments*

The experimental work was carried out in three columns made of polyacrylate sections. The column diameters were 0.1 m, 0.19 m and 0.38 m with total heights of 3 or 4 m. The 0.19 and 0.38 m diameter columns were equipped with sintered bronze plate gas distributor (with a mean pore size of 50 μm) and the 0.1 m diameter column with glass sintered plate distributor (mean pore size 150-200 μm). Two-stage cyclones were used to recover entrained fine particles and return these to the column. 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. A total of 1540 experiments were carried out with gas-solid systems with the following system properties and operating conditions:

*gas phase*: air ($\rho_G$=1. 3 kg/m$^3$); helium ($\rho_G$=0.18 kg/m$^3$); argon ($\rho_G$=1.83 kg/m$^3$); SF$_6$ ($\rho_G$=6.7 kg/m$^3$)

*solid phase*: fluidized cracking catalyst (FCC) ($\rho_{bulk}$ = 960 kg/m$^3$; $\rho_p$ = 1480 kg/m$^3$; particle size distribution: 10 % < 23 μm, 50 % < 49 μm, 90 % < 89 μm); polystyrene ($\rho_{bulk}$ = 608 kg/m$^3$; $\rho_p$ = 1073 kg/m$^3$; particle size distribution: 10 % < 480 μm, 50 % < 570 μm, 90 % < 630 μm)

*Operating conditions*: $U$ = 0.001 to 0.65 m/s; $H_0$ = 0.1 to 1.5 m.

### *Gas-liquid bubble column experiments*

The gas-liquid bubble column studies were carried out in columns of diameters 0.05, 0.1, 0.174, 0.19, 0.38 and 0.63 m. A typical experimental set-up is shown in Fig. 3 for the 0.63 m diameter bubble column. The column is made up of four polyacrylate sections with a total height of 4 m. The top of the column is connected to the exhaust. In all the experiments the pressure at the top of the column was nearly atmospheric. The 0.174, 0.19 and 0.38 m diameter columns were equipped with sintered bronze plate gas distributors (with a mean pore size of 50 μm). The 0.63 m diameter column was fitted with a spider shaped sparger; see Fig. 3. The 0.1 m diameter column was experimented with both a sintered glass plate distributor (with a mean pore size of 150-200 μm) and a polyacrylate sieve plate with 2.5 mm diameter holes. A total of 2878 experiments were carried out with gas-liquid systems with the following system properties and operating conditions.

*gas phase*: air; helium; argon; $SF_6$
*liquid phase*:

demineralized water ($\rho_L$ = 998 kg/m$^3$; $\mu_L$ = 0.001 Pa s; $\sigma$ = 0.072 N/m); paraffin oil (A) ($\rho_L$ = 795; $\mu_L$ = 0.023; $\sigma$ = 0.028); paraffin oil (B) ($\rho_L$ = 795; $\mu_L$ = 0.029; $\sigma$ = 0.028); tetradecane ($\rho_L$ = 763; $\mu_L$ = 0.022; $\sigma$ = 0.027); aqueous polyacrylamide solutions of concentration 100, 250, 500 and 1000 ppm wt ($\rho_L$ = 998).

*Operating conditions*: $U$ = 0.001 to 0.891 m/s; $H_0$ = 0.3 to 2.2 m

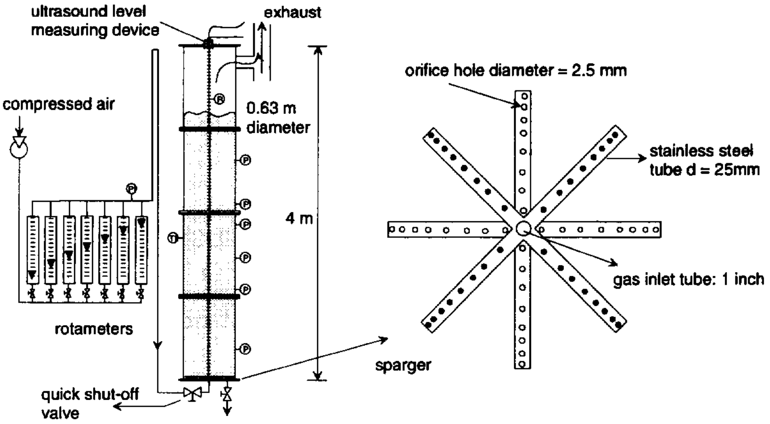

Fig. 3. Experimental set up for 0.63 m diameter bubble column fitted with spider shaped sparger.

***Bed collapse experiments***

For characterising the hydrodynamics of G-S and G-L systems, dynamic gas disengagement i.e. bed collapse experiments were performed. The dispersion height was monitored visually and also by means of a pressure sensor connected to a PC for continuous recording of the voltage signals (these are proportional to the hydrostatic head). This technique is well known and described in the literature (Daly *et al.*, 1992; Schumpe and Grund, 1986). Figure 4 shows typical bed collapse experiments with air-FCC and air-tetradecane. The initial sharp decrease in height is due to escape of the "dilute" phase gas (= "bubbles" in G-S fluid beds; = fast-rising "large" bubbles in G-L bubble columns; this is followed by slow disengagement of the gas entrapped in the "dense" phase (= "emulsion" gas in G-S fluid beds; = "small" bubbles in G-L bubble columns). It is noted from Fig. 4 that the slope of the initial collapse is approximately equal for G-S and G-L systems; this implies that the dilute phase rise velocities are approximately of the same order of magnitude. The slope of the second, slowly disengaging, portion of the collapse curves in Fig. 4 yields the superficial gas velocity through the dense phase, $U_{df}$. The "bubbles" in G-S fluid bed and "large" bubbles in G-L bubbles are considered to be equivalent. For G-L bubble columns, the "small" bubbles are typically 2 - 5 mm in diameter and their holdup is strongly dependent on the physical properties of the system. From the collapse curves, the following parameters can be determined. The *total gas voidage*, or hold-up for G-S fluid bed was calculated from $\varepsilon = (H - \rho_{bulk} H_0/\rho_p)/H$. For G-L bubble columns the total gas voidage from $\varepsilon = (H - H_0)/H$. The *gas hold-up of the "dilute" phase*, $\varepsilon_b$, is determined from $\varepsilon_b = (H - H_1)/H$. The *gas voidage in the "dense" phase* is $\varepsilon_{df} = (\varepsilon - \varepsilon_b)/(1 - \varepsilon_b)$. On the basis of the few thousand collapse experiments with various systems an analogous picture of the hydrodynamics emerged, as discussed in the following sections.

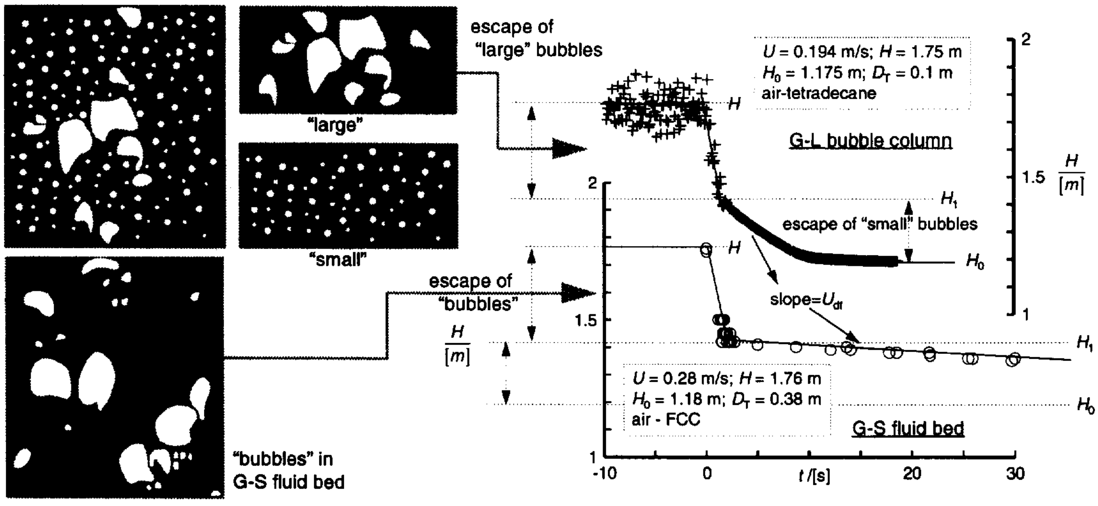

Fig. 4. Dynamic gas disengagement experiments in G-S fluid beds and G-L bubble columns. The initial sharp disengagement is due to the escape of the "dilute" phase (= "bubbles" in G-S = "large" bubbles in G-L). The second, slower disengagement is due to escape of gas in the "dense" phase (= "emulsion" gas in G-S = "small" bubbles in G-L). An artist's impression, based on flow visualization in a 2D column, of the dilute phase in G-L and G-S systems, is shown on the left.

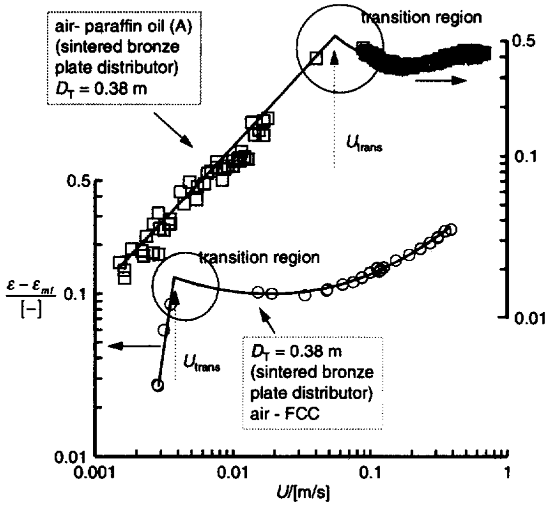

Fig. 5. Total gas voidage in G-S fluid beds and G-L bubble columns.

## TOTAL AND DENSE PHASE GAS VOIDAGES

For G-S fluid beds, the bed expands only when the minimum fluidization velocity $U_{mf}$ is exceeded. For G-L systems we have $U_{mf} = 0$; $\varepsilon_{mf} = 0$. In Fig. 5 we compare ($\varepsilon$ - $\varepsilon_{mf}$) for fluid beds with $\varepsilon$ for G-L systems. The initial, linear, portion of the Voidage - $U$ curve represents the homogeneous regime. Both curves exhibit a maximum at the regime transition point, $U_{trans}$; this maximum is prominent for columns with sintered plate distributor. For G-S fluid beds $U_{trans}$ corresponds to the minimum bubbling point $U_{mb}$. For sieve plates and spargers the maximum at $U_{trans}$ is not observed. Beyond the transition point (region), we have the heterogeneous regime. The regime transition point $U_{trans}$ is shifted to higher values with increasing gas density for both G-S and G-L systems; in other words increasing pressure has a stabilising effect on the homogeneous flow regime (Krishna *et al.*, 1993; Wilkinson *et al.*, 1992). Increasing particle size in fluid beds leads to earlier transition; for fluid bed experiments with polystyrene no homogeneous expansion was observed. This destabilising effect is also observed for G-L systems with increasing liquid viscosity (Stegeman, 1994) and the behaviour corresponds to fluid beds with Geldart B type powders (Geldart, 1986).

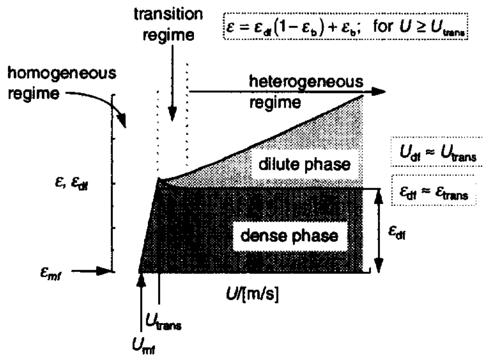

Fig. 6. Generalized model for gas holdup.

The dense phase gas voidage $\varepsilon_{df}$ was found to be practically constant in the heterogeneous regime; a common picture for the gas voidages in G-S and G-L systems which emerged from the experiments is shown in Fig. 6. For estimation purposes we may take $\varepsilon_{df} \approx \varepsilon_{trans}$ and $U_{df} \approx U_{trans}$; the transition parameters may be estimated for G-S systems from the correlations in Geldart (1986) and for G-L columns using the correlations of Reilly *et al.* (1994) or Wilkinson *et al.* (1992). The dense phase gas voidage $\varepsilon_{df}$ is significantly affected by system physical properties but is relatively insensitive to the scale of operation; this parameter can therefore be determined in a column of relatively small scale under realistic conditions of temperature and pressure.

## DILUTE PHASE GAS HOLDUP

The rise velocity of the dilute phase in fluid beds is known to be a strong function of the scale of operation and the Werther (1983) correlation assumes the form $V_b = \phi \sqrt{(g\, d_b)}$; $\phi = \phi_0\, D_T^{\,N}$ . The bubble size in fluid beds can be calculated from the Darton *et al.* (1977) relationship $d_b = \alpha_1\, (U - U_{df})^{2/5}\, (h + h_0)^{4/5}\, g^{-1/5}$ where the constant $\alpha_1$ = 0.54. The bubble growth in fluid beds of fine powders ($d_p$ < 100 μm) is restricted to an equilibrium size $d_b$* and the corresponding height above the distributor where the bubbles reach their equilibrium size $d_b$* is $h$*. Ellenberger and Krishna (1994) combined the Darton bubble growth model with the Werther type rise velocity relationship to obtain an analytic expression for the dilute phase gas holdup $\varepsilon_b$ :

$$\varepsilon_b = \frac{1}{H}\int_0^H \frac{\left(U-U_{df}\right)}{V_b} =$$

$$= \frac{1}{\sqrt{\alpha_1}\;\phi_0\, D_T^N\; g^{2/5}} \frac{\left[\left(h^*+h_0\right)^{3/5} - \left(h_0\right)^{3/5}\right]}{\left(3/5\right)} \frac{\left(U-U_{df}\right)^{4/5}}{H} + \frac{1}{\sqrt{\alpha_1}\;\phi_0\, D_T^N\; g^{2/5}}\left(h^*+h_0\right)^{-2/5}\left(H-h^*\right)\frac{\left(U-U_{df}\right)^{4/5}}{H} \tag{1}$$

For air-FCC the experimental data for $\varepsilon_b$ as a function of ($U$ - $U_{df}$), $H$ and $D_T$ can be fitted to obtain the model parameters in eq. (1). These results, presented earlier by Ellenberger and Krishna (1994), are $\phi$ = 1.95 $D_T^{\,1/3}$ with the equilibration height fitted with either a linear, $h^* = 0.33 + 1.51\ (U - U_{df})$, or a power law, $h^* = 1.16\ (U - U_{df})^{0.33}$, relation. The equilibration height $h$* lies in the range 0.2 - 1.2 m for industrial operations. For fast reactions such as catalytic partial oxidation of natural gas for syngas production the proper description of bubble growth in the initial bubble growth zone is of vital importance, as we shall demonstrate below. If we are just interested in determining the dilute phase gas holdup in tall beds, say taller than 1.5 m it is possible to simplify eq. (1) to obtain for $H >> h^*$:

$$\varepsilon_b = \frac{1}{\sqrt{\alpha_1}\,\phi_0\, D_T^N\, g^{2/5}} \left(h^* + h_0\right)^{-2/5} \left(U - U_{df}\right)^{4/5} \quad \text{for } H >> h^* \tag{2}$$

Assuming a power law dependence: $h^* = b0\,(U - U_{df})^{b1}$, we obtain after neglecting $h_0$ in comparison to $h^*$:

$$\varepsilon_b = \alpha_2 \frac{1}{D_T^N} \frac{1}{\left(U - U_{df}\right)^{b2}} \left(U - U_{df}\right)^{4/5}; \quad \alpha_2 = \frac{1}{\sqrt{\alpha_1}\,\phi_0\, g^{2/5}} (b0)^{-2/5}; \quad b2 = \frac{2}{5} b1 \tag{3}$$

The parameters of the simplified model in eq. (3) are $\alpha_2$, $N$, and $b2$. By re-fitting air-FCC data for bed heights $H$ in excess of 1.5 m, and for $U > 0.1$ m/s we obtain $\alpha_2 = 0.287$; $N = 0.333$ and $b2 = 0.175$. Figure 7 which compares the measured data with the model fits for three column diameters testifies to the importance of scale effects.

For G-L bubble columns the equilibration height $h^*$ is of the order of 0.1 - 0.3 m (Ellenberger and Krishna, 1994), much smaller than for G-S fluid beds, and for dispersion heights greater than 1 m the requirement $H >> h^*$ is easily met. Therefore eq. (3) can be expected to apply here. Further, from a detailed analysis, set by set, of the 2878 experiments in G-L bubble columns we established that $\varepsilon_b$ (= gas holdup of "large" bubbles), for $U > 0.1$ m/s, is: (1) virtually independent of liquid properties, (2) virtually independent of the manner in which the gas is distributed, and (3) independent of the density of the gas phase. We fitted the model parameters $\alpha_2$, $N$ and $b2$ selecting only experimental dilute phase gas holdup $\varepsilon_b$ vs $(U - U_{df})$ data values corresponding to dispersion heights $H > 1$ m and for $U > 0.1$ m/s. The fit parameter parameters thus obtained are $\alpha_2 = 0.268$; $N = 0.18$ and $b2 = 0.22$, which are remarkably close to the corresponding fluid bed parameters. The column diameter dependence of the dilute phase gas holdup in bubble columns is somewhat weaker than for fluid beds, but this effect cannot be neglected in scaling up to large diameter columns. Figure 7 also presents a comparison of the experimental data for air-paraffin oil (A) for three column diameters with the model calculations of eq. (3). The quantitative analogies between G-S and G-L systems are evident from Fig. 7.

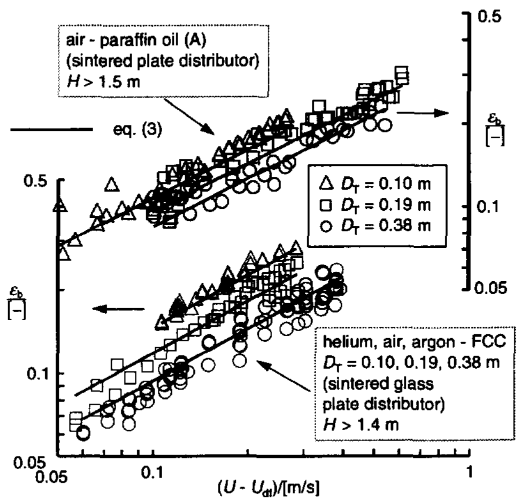

Fig. 7. Dilute phase gas holdup in G-S fluid beds and G-L bubble columns.

The dilute phase gas holdup is hardly affected by the gas density within the range of gas densities studied, even though the total gas voidage is increased with increased $\rho_G$; see Fig. 8. Again the analogies with respect to the influence of $\rho_G$ is evident. Also drawn in Fig. 8 (b) are the predictions of the "large" bubble gas holdup for He and $SF_6$ using the Wilkinson *et al.* (1992) correlation; this

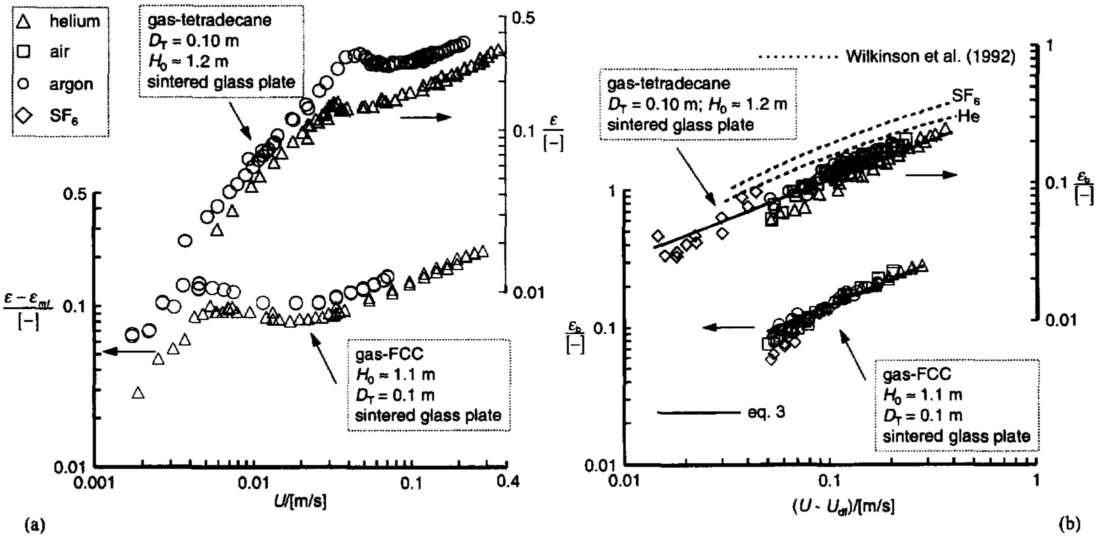

Fig. 8. Influence of gas density on (a) total gas voidage and (b) dilute phase gas holdup. $U_{df}$ assumed to be equal to $U_{trans}$.

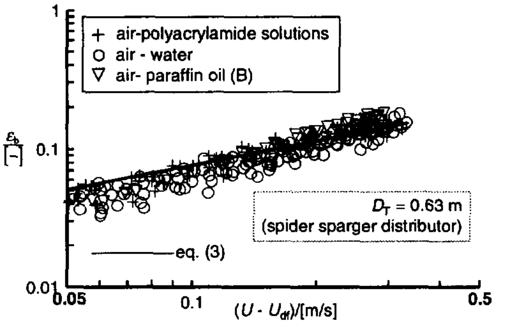

Fig. 9. Influence of liquid properties on the dilute phase gas holdup in G-L bubble columns.

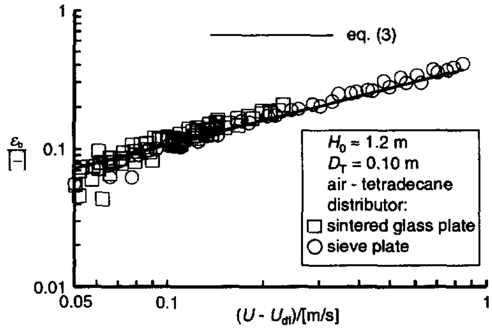

Fig. 10. Influence of gas distributor type on the dilute phase gas holdup in G-L bubble columns.

correlation significantly overpredicts $\varepsilon_b$ and anticipates, incorrectly, a fairly significant effect of $\rho_G$ on $\varepsilon_b$. Equation (3) is much simpler and far more accurate than the Wilkinson correlation for $\varepsilon_b$.

A further drawback of the Wilkinson correlation is that it predicts that $\varepsilon_b$ is also dependent on the physical properties of the liquid ($\rho_L$, $\mu_L$, $\sigma$); our experimental data show that $\varepsilon_b$ is practically independent of these properties; cf. Fig. 9. Measurements with air-tetradecane in a 0.1 m diameter column show that the manner in which gas is distributed into the column has a negligible influence on the dilute phase gas holdup; see Fig. 10. We may expect an analogous gas distributor effect in G-S fluid beds.

## HOLDUPS IN SLURRY BUBBLE COLUMNS

Slurry bubble column reactors, used in Fischer Tropsch and methanol synthesis (cf. Fig. 1), usually employ fairly fine catalyst particles, typically smaller than 100 μm. Such small particles are finely dispersed in the liquid phase and will have the mixing characteristics of the liquid phase. Here the "dense" phase can be considered to be the slurry in which the "small" bubbles are dispersed. As in G-L systems, in the heterogeneous regime the dilute phase consisting of "large" bubbles traverse the column at high velocities. De Swart and Krishna (1995) studied the influence of particles concentration on the gas holdup in a slurry bubble column of 0.05 m diameter. With increasing slurry concentration the total gas holdup $\varepsilon$ is found to decrease significantly and regime transition occurs "earlier"; see Fig. 11 (a). The "dilute" phase gas hold-up $\varepsilon_b$ on the other hand is practically independent of the slurry concentration; cf Fig. 11 (b). The decrease in $\varepsilon$ is almost entirely to be attributed to the decrease in the gas voidage of the "small" bubbles, $\varepsilon_{df}$. Solid particles enhance the coalescence of "small" bubbles but have no significant effect on the fast-rising "large" bubbles.

An acid test of the model developed above for estimation of the dilute phase gas holdup, $\varepsilon_b$, is to apply eq. (3) to try to predict $\varepsilon_b$ for slurry systems. A comparison of eq. (3) predictions, using $D_T$ = 0.05 m with experimental data, shown in Fig. 11 (b), testifies to the predictive (in this case the scale-down) capability of the model. For estimation of the gas holdup of commercial slurry reactors, it is recommended that $\varepsilon_{df}$ be measured in a small diameter column with the reaction mixture under actual conditions of temperature and pressure. Since it is also to be expected that the dense phase gas holdup is dependent on the type of gas distribution, these small scale hot experiments need to be conducted in an equipment with commercial type distribution devices. The total gas holdup is then estimated from $\varepsilon = \varepsilon_b + (1 - \varepsilon_b)\,\varepsilon_{df}$. The advantage of this approach is that we separately estimate the scale-

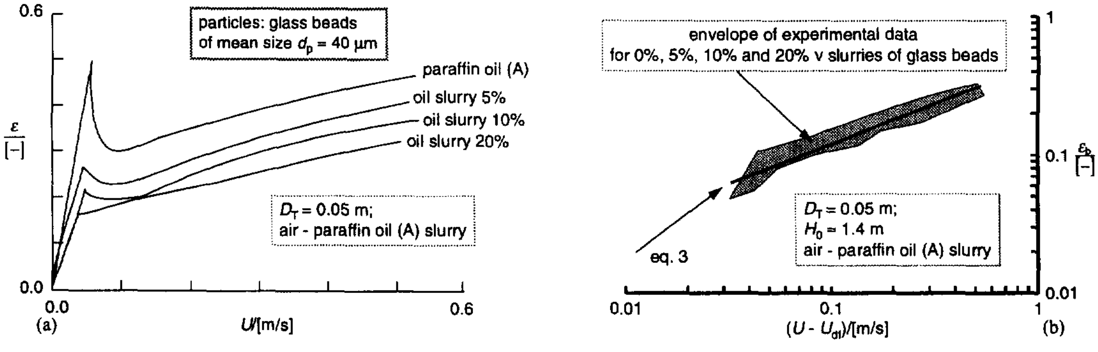

Fig. 11. Influence of slurry concentration on (a) total gas holdup and (b) dilute phase gas holdup. (a) shows a significant lowering of the total gas holdup with increased slurry concentration. (b) shows that the "large" bubble gas holdup is virtually unaffected. The line drawn in Figure 11 (b) is with eq. (3) using parameters fitted for *liquids*. Data scanned from De Swart and Krishna (1995).

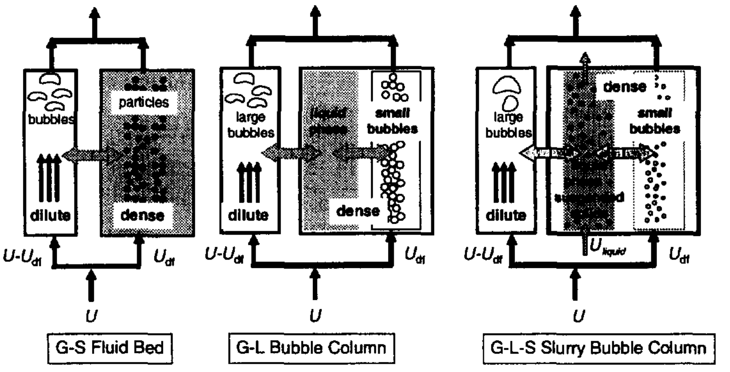

Fig. 12. Generalised "two-phase" model.

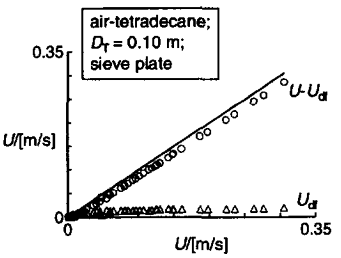

Fig. 13. Superficial gas velocities through "dilute" and "dense" phases in G-L bubble column with air-tetradecane.

dependent, system-property-independent $\varepsilon_b$ from the scale-independent, system-property-dependent parameter $\varepsilon_{df}$. The total gas holdup $\varepsilon$ is dependent in a complicated manner on system properties *and* on scale.

## GENERALISED TWO-PHASE MODEL FOR REACTOR CONVERSION CALCULATIONS

Based on the common hydrodynamic picture which has emerged from our experimental studies we suggest a generalisation of the "two-phase" model of May (1959) and Van Deemter (1961), developed for G-S fluid beds, to G-L and G-L-S bubble columns; this extension is pictured in Fig. 12. The "dilute" phase is to be identified with the solids-free bubbles in a fluid bed or the fast-rising large bubble population in G-L, G-L-S bubble columns; this phase travels up the column virtually in plug flow. The "dense" phase in a fluid bed consists of the suspension of solids with a gas flow corresponding to $U_{df}$. For bubble columns the dense phase is to be identified with the liquid phase *together with* the small bubbles which are entrained in the liquid. For G-L-S slurry columns the "dense" phase is identified with the liquid phase along with the catalyst particles and the entrained "small" bubbles. In the heterogeneous flow regime, the small bubbles have the backmixing characteristics of the liquid, or slurry, phase.

In G-S fluid beds with fine particles (< 100 μm), $U_{df}$ is in the range of 0.005 - 0.01 m/s and for typical industrial operations with $U$ > 0.3 m/s, the gas phase conversion will be dictated by mass transfer from the dilute phase. A similar picture also holds for G-L bubble columns and is illustrated in Fig. 13 with the experimental data for air-tetradecane. For bubble column slurry reactors, the "small" bubble population is virtually destroyed by use of high slurry concentrations and the gas phase conversion will be dictated by mass transfer from "large" fast-rising bubbles (De Swart and Krishna, 1995).

## MASS TRANSFER FROM THE DILUTE PHASE

For G-S fluid beds the Davidson *et al.*(1977) model can be used to calculate the mass transfer coefficient per unit volume of the dilute phase $k_G\,a' = 7.14\,(U_{df}/d_b) + 5.46\,D_G^{1/2}\,(\varepsilon_{df}/(1+\varepsilon_{df}))/d_b^{5/4}$. The number of transfer units, *NTU*, also called the cross-flow ratio, can be calculated by combining the Davidson mass transfer model with the Darton bubble growth model along with the rise velocity relationship $V_b = \phi_0\,D_T^{N}\sqrt{(g\,d_b)}$.

$$NTU = \int_0^H \frac{k_g a'}{V_b}\,\mathrm{d}h = \int_0^{h^*} \frac{k_g a'}{V_b}\,\mathrm{d}h + \int_{h^*}^{H} \frac{k_g a'}{V_b}\,\mathrm{d}h \qquad (4)$$

The integration in the region 0 - $h^*$ has to account for bubble growth. To demonstrate the importance of scale

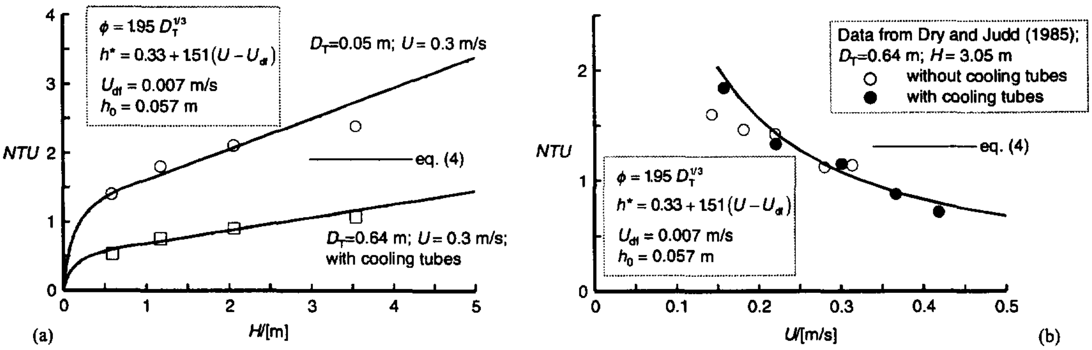

Fig. 14. Comparison of NTU calculations with eq. (4) with experimental data of Dry and Judd (1985). (a) shows the axial profiles of *NTU*. (b) shows the influence of increasing superficial gas velocity on *NTU* for fluid beds with and without vertical cooling tubes inserted into them.

effects we simulated the results of Dry and Judd (1985) presented in Figs 11 and 13 of their paper. Comparison of calculations with eq. (4), with parameters $\phi = 1.95\, D_T^{1/3}$ and $h^* = 0.33 + 1.51\,(U - U_{df})$ are shown in Figs 14 (a) and (b). The values of $h_0$, characterising the distributor, and $U_{df}$ were not reported by Dry and Judd and the parameter values had to be assumed. Even though assumed values of $h_0$ and $U_{df}$ were used, the model simulations, using eq. (4), can be considered to be remarkably good and the right trends with increasing $U$ and with increasing scale ($H$ and $D_T$) are reproduced. An important experimental result shown in Fig. 14 (b) is that the influence of vertical cooling tubes on *NTU* is negligibly small. In other words vertical cooling tubes do not tend to break up bubbles; this has important consequences in scaling up to commercial units.

There are no corresponding experimental studies reported on mass transfer from "large" bubbles in bubble column slurry reactors operating under non-slugging conditions at high superficial gas velocities say 0.3 m/s, typical of industrial operations for the Fischer Tropsch process. On the basis of the information available for G-S fluid beds, and using analogies as a conceptual tool, we could anticipate that mass transfer in the Fischer Tropsch slurry reactor would be relatively insensitive to the presence of vertical cooling tubes. This conclusion requires to be verified experimentally.

## BACKMIXING OF THE DENSE PHASE

The fast-rising "dilute" phase tend to churn up the "dense" phase as pictured in Fig. 15. It may be expected that the backmixing coefficient must be relatable to the rise velocity of the fast rising large bubbles, $V_b$ and to the size of the eddies in the liquid phase generated by these fast-rising bubbles. The maximum size of the eddies, for vessels with $H/D_T > 1$, is limited by the column diameter $D_T$. Drawing inspiration from the unified approach to axial mixing in multiphase contactors of Joshi (1980) we may anticipate that the dense phase axial dispersion coefficient is determined by the product of $V_b$ and $D_T$, i.e. $D_{ax} = \alpha_3 V_b\, D_T$, where $\alpha_3$ is a constant which must be determined by experiment. For dispersion heights $H > 1$ m and for $H/D_T > 1$ we may estimate the dilute phase rise velocity using eq. (3):

$$V_b \equiv \frac{U - U_{df}}{\varepsilon_b} = \frac{1}{\alpha_2}\left(U - U_{df}\right)^{b2+\frac{1}{5}} D_T^N \qquad (5)$$

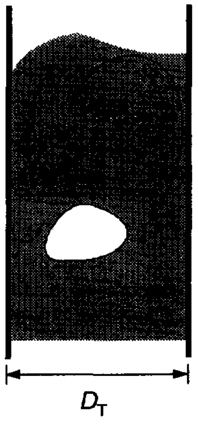

Fig. 15. The fast rising "dilute" phase churns up the "dense" phase.

The maximum column diameter used in our experiment was 0.63 m and the question of extrapolation to commercial scale reactors arises. The Werther (1983) correlation of experimental rise velocity data for fluid beds shows that the rise velocity constant $\phi$ remains constant for diameters above 1 m. There is no corresponding data for bubble columns and, again relying on hydrodynamic analogies, we can assume a similar plateau in the rise velocity constant above 1 m diameter. This means that eq. (5) can be used for $D_T > 1$ m by setting $D_T = 1$ m.

It is interesting to note that taking $\alpha_3 = 0.25$, i.e. with $D_{ax} = 0.25\, V_b\, D_T$ the calculations of the axial dispersion coefficient calculations closely correspond to those of the Baird and Rice (1975) correlation: $D_{ax} = 0.35\,(gU)^{1/3}\, D_T^{4/3}$. It appears that we do not need to correlate $D_{ax}$ parameter separately, because we can derive it "naturally" from the already established analogies in the hydrodynamics of G-S and G-S systems.

There is a lot of information on backmixing of the dense phase in G-S fluid beds (e.g. Van Deemter, 1980) and in view of the hydrodynamic analogies, now established, this data can be assumed to be valid for G-L-S slurry reactors for which there is almost no information available on large scale columns.

## CONCLUSIONS

In this paper we have put forward a scale-up procedure for G-S fluid beds and G-L-(S) bubble column reactors relying on their analogous hydrodynamic behaviour. These analogies are summarised below.

(1) The regime transition velocity $U_{trans}$ and gas holdup $\varepsilon_{trans}$ are both increased with increasing gas density.

(2) Increased particle size in G-S fluid beds leads to "earlier" transition. An analogous effect is found in G-L bubble columns with increasing liquid viscosity or slurry concentration.

(3) The hydrodynamics of G-S fluid beds and G-L bubble columns can be modelled most conveniently using a generalised two-phase model pictured in Fig. 12.

(4) A generalised picture for gas voidage in G-S fluid beds and G-L bubble columns has been established as shown in Fig. 6. The superficial gas velocity through the dense phase $U_{df}$ is approximately equal to the velocity at the regime transition point, $U_{trans}$. The gas voidage in the dense phase $\varepsilon_{df}$ is approximately equal to gas voidage at the regime transition point, $\varepsilon_{trans}$; this parameter is affected in a significant manner by the system properties (gas density, either particle size and size distribution or liquid viscosity and surface tension). The dense phase gas voidage is practically independent of the superficial gas velocity for $U > 0.1$ m/s.

(5) For both G-S fluid beds and G-L bubble columns the gas holdup of the dilute phase can be calculated on the basis of a model based on bubble growth, eq. (1). The equilibration height $h^*$ is 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 gas-liquid systems. As a consequence the influence of the dispersion height $H$ on the dilute phase holdup is stronger for air-FCC than for gas-liquid bubble columns. For large dispersion heights say greater than 1.5 m, the simplified model given in eq. (3) applies equally to G-S fluid beds and G-L bubble columns.

(6) The rise velocity of the dilute phase $V_b$ for both fluid beds and bubble columns increases with column diameter; this dependence is quantified by the exponent $N$. The fitted value of the exponent for air-FCC is 0.333 whereas for all gas-liquid systems the exponent has been determined to be 0.18. This means that the diameter effect on the dilute phase gas holdup is weaker for G-L bubble columns than it is for G-S fluid beds. The rise velocity of the dilute phase can be calculated by eq. (5).

(7) In bubble column operation, the influence of liquid properties on the dilute phase gas holdup in insignificant. Also, the manner in which gas is distributed in the column has little influence on the dilute phase gas holdup.

(8) The influence of gas density on the dilute phase gas holdup is insignificant for both fluid beds and bubble columns. This conclusion must be viewed with caution in view of the fact that the variation of gas density in our own experiments has only been up to 6.7 kg/m$^3$. There is need for measurements under high pressure conditions in G-S and G-L systems.

(9) The axial dispersion coefficient for the dense phase $D_{ax}$ is simply calculable from $D_{ax} = 0.25\ V_b D_T$.

Mass transfer from "large" bubbles in slurry bubble columns has not received due attention in the literature. Vermeer and Krishna (1981), in a study of mass transfer from large bubbles in a G-L bubble column found that the values of mass transfer coefficient per unit "large" bubble holdup $k_L\, a'$ were much higher than what one might expect from bubble sizes in the range of 0.05 - 0.08 m. In view of the several hydrodynamic analogies we have uncovered in the present study, we could wonder if these high $k_L\, a'$ values are to be attributable to frequent coalescence and splitting mechanisms of "large" bubble population, a mechanism which has been verified to be true for the mass transfer from the dilute phase in G-S fluid beds (Sit and Grace, 1981). If such a mechanism is found to be true for G-L system, this would be yet another demonstration of the power of our suggested analogous approach.

## NOTATION

| | |
|---|---|
| $a'$ | interfacial area per unit volume of dilute phase, m$^{-1}$ |
| $b0, b1$ | fit parameters in relations for equilibration height $h^* = b0\,(U - U_{df})^{b1}$ |
| $b2$ | fit parameter in unified dilute phase gas holdup model eq. (3) |
| $d_h$ | bubble diameter of dilute phase, m |
| $d_b^*$ | equilibrium bubble size of dilute phase, m |
| $d_p$ | mean particle size, m |
| $D_{ax}$ | axial dispersion coefficient, m$^2$ s$^{-1}$ |
| $D_G$ | gas phase molecular diffusion coefficient, m$^2$ s$^{-1}$ |
| $D_T$ | column diameter, m |
| $g$ | acceleration due to gravity, 9.81 m s$^{-2}$ |
| $h$ | height above the gas distributor, m |
| $h^*$ | height above the gas distributor where the bubbles reach equilibrium, m |
| $h_0$ | parameter determining the initial bubble size ($h_0$ = 0.03 m for porous plate distributors), m |
| $H, H_0, H_1$ | height of expanded bed, ungassed bed and after escape of dilute phase, m |
| $k_G a'$ | mass transfer coefficient per unit volume of dilute phase for G-S fluid bed, s$^{-1}$ |
| $k_L a'$ | liquid phase mass transfer coefficient per unit volume of dilute phase in G-L system, s$^{-1}$ |
| $N$ | exponent in the rise velocity relationship $V_b = \phi \sqrt{(g\, d_b)}$; $\phi = \phi_0\, D_T^N$ |
| $NTU$ | number of mass transfer units for transfer from the dilute phase |
| $t$ | time, s |
| $U$ | superficial gas velocity, m s$^{-1}$ |
| $(U - U_{df})$ | superficial gas velocity through the dilute phase, m s$^{-1}$ |
| $U_{df}$ | superficial velocity of gas through the dense phase, m s$^{-1}$ |
| $U_{mb}$ | superficial velocity at which the first "bubbles" are formed, m s$^{-1}$ |
| $U_{mf}$ | minimum fluidization velocity (corresponds to $U_{trans}$,), m s$^{-1}$ |
| $U_{trans}$ | superficial gas velocity at regime transition, m s$^{-1}$ |
| $V_b$ | rise velocity of the dilute phase, m s$^{-1}$ |
| $V_{circ,dense}$ | circulation velocity of the dense phase, m s$^{-1}$ |

### *Greek letters*

| | |
|---|---|
| $\alpha_1$ | constant in Darton bubble growth model |
| $\alpha_2$ | parameter in the simplfied model for dilute phase gas holdup, eq. (3) |
| $\alpha_3$ | parameter in model for dense phase backmixing |

| | |
|---|---|
| $\varepsilon$ | total gas voidage of G-S or G-L system |
| $\varepsilon_b$ | gas hold-up of "dilute" phase; = "bubbles" in G-S systems; = "large" bubbles in G-L systems |
| $\varepsilon_{df}$ | hold-up of gas in "dense" phase; corresponds to "small" bubble gas holdup in G-L systems |
| $\varepsilon_{mf}$ | voidage of G-S fluidized bed at minimum fluidization conditions |
| $\varepsilon_{trans}$ | gas hold-up at the regime transition point |
| $\mu_G$, $\mu_L$ | viscosity of gas and liquid phases, Pa s |
| $\rho_{bulk}$, $\rho_p$ | bulk and particle densities, kg $m^{-3}$ |
| $\rho_G$, $\rho_L$ | density of gaseous and liquid phases, kg $m^{-3}$ |
| $\sigma$ | surface tension of liquid phase, N $m^{-1}$ |
| $\phi$, $\phi_0$ | constants in the rise velocity relationship for the dilute phase $V_b = \phi \sqrt{(g\, d_b)}$; $\phi = \phi_0\, D_T^{N}$ |

***Subscripts***

| | |
|---|---|
| b, df | referring to "dilute" and "dense" phases respectively |
| G, L | referring to gas and liquid phases |
| trans | referring to the transition point |
| T | tower or column |

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