IChemE

0263-8762/95/$07.00+0.00

INFLUENCE OF PARTICLES CONCENTRATION ON THE HYDRODYNAMICS OF BUBBLE COLUMN SLURRY REACTORS

J. W. A. DE SWART (GRADUATE) and R. KRISHNA (FELLOW)

Department of Chemical Engineering, University of Amsterdam, The Netherlands

This study focuses on the influence of particles concentration on the hydrodynamics of bubble column slurry reactors operating in the heterogeneous flow regime. Experiments were carried out in a 50 mm diameter glass column with paraffin oil as the liquid phase and glass beads of 40 μm diameter as the solids phase. The particles concentrations studied were 5, 10 and 20% v. For interpretation of the experimental results a generalization of the 'two-phase' model for gas-solid fluid beds was used to model the bubble hydrodynamics. The two phases are identified as follows: (i) a 'dilute' phase consisting of the fast-rising 'large' bubbles which traverse the column virtually in plug flow, and (ii) a 'dense' phase which is identified with the liquid phase along with the solid particles and the entrained 'small' bubbles. The 'dense' phase suffers a considerable degree of backmixing. Dynamic gas disengagement experiments were carried out in the heterogeneous flow regime to determine the gas voidage in the 'dilute' and 'dense' phases.

The experimental data show that increasing the solids concentration results in a pronounced decrease in the total gas hold-up but the influence on the 'dilute' phase gas hold-up is negligible. The 'dense' phase gas voidage suffers a significant decrease in gas hold-up due to enhanced coalescence of the 'small' bubbles resulting from introduction of particles. The virtual independence of the 'dilute' phase hold-up on the liquid phase properties was confirmed by measurements with ethanol, octanol, water and aqueous NaOH solutions. The 'dilute' phase gas hold-up could be described using a bubble growth model which accounts for the influence of the column diameter and column height.

Keywords: bubble columns; dense phase; dilute phase; fluidized beds; homogeneous regime; heterogeneous regime; hydrodynamics; multiphase reactors; particles; scale up; slurry reactors

INTRODUCTION

The overall aim of our investigation is to develop a fundamentally based scale up procedure for bubble column slurry reactors, which find application in the chemical industry in processes such hydrogenations and oxidations[1]. The Fischer-Tropsch synthesis of hydrocarbons from syngas is another important, emerging, application of this reactor type[2]. The present study distinguishes itself from earlier studies on the hydrodynamics of bubble column slurry reactors (see e.g. Bukur *et al.*,[3,4]; Deckwer *et al.*,[5–7]; Fukuma *et al.*,[8]; Kara *et al.*,[9]; Kelkar *et al.*,[10]; Koide *et al.*[11]; O'Dowd *et al.*,[12]; Saxena *et al.*,[13–15]; Schumpe *et al.*,[16,17]; Shah *et al.*,[18]; Shetty *et al.*,[19]) in that the focus is on the influence of increased particles concentration on the gas holdups in the 'large' and 'small' bubble populations in the heterogeneous flow regime. For the interpretation of our experimental results in the heterogeneous flow regime we adopt the generalization of the two-phase model of Van Deemter[20] and May[21], developed for gas-solid fluidized beds; see Figure 1. The 'dilute' phase is identified with the fast-rising 'large' bubbles which traverse the column virtually in plug flow. The 'dense' phase is identified with the liquid phase along with the solid particles and the entrained 'small' bubbles. The 'dense' phase suffers a considerable degree of backmixing. The influence of particles concentration has been studied on the gas voidage (i.e. hold-up) of both the 'dilute' and 'dense' phases. The model pictured in Figure 1 is an extension the two-bubble class model suggested in the literature[18,22].

EXPERIMENTAL

Most of the experiments were performed in a 50 mm i.d. column of 4.5 m height. A paraffinic mineral oil (viscosity $\mu_L = 2.4$ mPa s; density $\rho_L = 796$ kg/m^3; surface tension $\sigma = 28$ mN/m) was used as the liquid phase, air as gaseous phase and spherical glass beads with a mean particle size of 40 μm formed the suspended solids phase. Experiments 'small' bubble classes were determined by means of dynamic gas disengagement experiments using a pressure transducer, as described in the literature[19,23]. Gas distribution was by means of a sintered glass distributor having a mean pore size of 200 mm. The experimental set-up is shown schematically in Figure 2.

Experiments were also carried out in the 50 mm i.d. column with ethanol ($\mu_L = 1.2$ mPa s; $\rho_L = 789$ kg/m^3; $\sigma = 23$ mN/m) and octanol ($\mu_L = 8.87$ mPa s; $\rho_L = 827$ kg/m^3; $\sigma = 27.5$ mN/m) in order to study the influence of liquid properties on bubble hydrodynamics. A few experiments were carried out with demineralized water ($\mu_L = 1$ mPa s; $\rho_L = 998$ kg/m^3; $\sigma = 72$ mN/m) and with aqeuous solutions of NaOH in a 0.174 m diameter column of 3 m height to study the influence of electrolytes and of column diameter on bubble hydrodynamics.

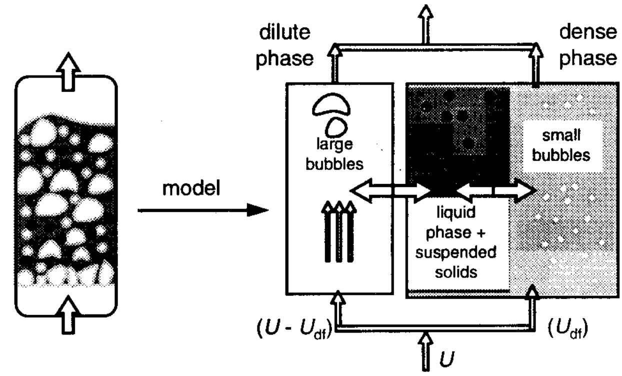

Figure 1. Generalized two-phase model applied to a bubble column slurry reactor.

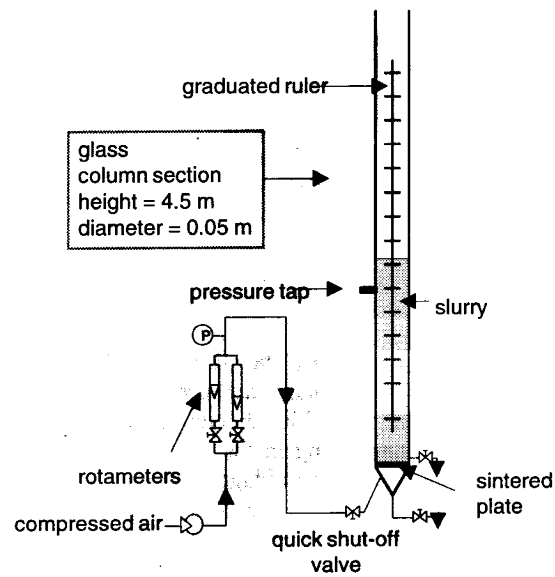

Figure 2. Schematic of experimental set-up.

RESULTS AND DISCUSSION

Typical dynamic gas disengagement experiments for paraffin oil and paraffin oil containing 20% volume of glass beds are shown, respectively, in Figures 3 and 4. The ungassed dispersion height is denoted by H_0 and the dispersion height of the gas-liquid (+ solids) is denoted by H. The total gas voidage is thus determined from

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

When the gas is switched off instantaneously by means of a quick shut-off valve, the bed height decreases sharply due to escape of the 'large' fast-rising bubbles. The gas hold-up of the 'dilute' phase is determined from

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

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 the paraffin oil severe foaming tendency was observed. One could distinguish between 'small' and 'micro' bubble disengagement regimes. The 'micro' bubbles are typically smaller than about 1 mm in diameter. For the purposes of the analysis in this paper, which largely focuses on the 'large' bubble, the 'small' and 'micro' bubble populations are lumped into one population which is termed the 'dense' phase gas. Thus, the hold-up of the gas in the 'dense' phase was determined using

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

It is important to note here that the definition of 'small' bubble holdup in bubble columns in the literature[6] is different to the one used in equation (3) for the 'dense' phase gas voidage. The slope of the disengagement curve

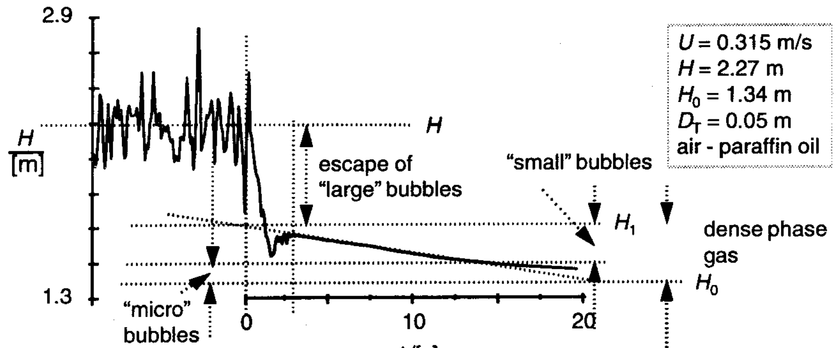

Figure 3. Typical dynamic gas disengagement experiment with air-paraffin oil.

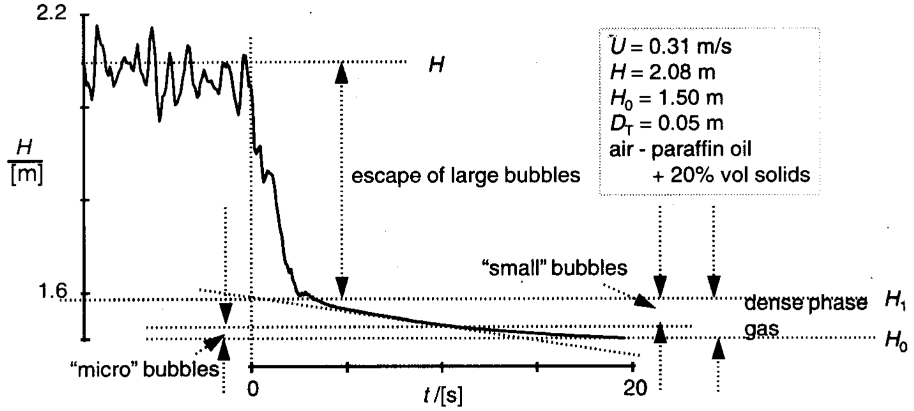

Figure 4. Typical dynamic gas disengagement experiment with air–20% paraffin oil slurry.

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 superficial gas velocity through the 'micro' bubbles is of the order of 0.5 mm/s, much smaller than the superficial gas velocity through the 'small' bubbles.

The total gas holdup is found to decrease significantly with increasing slurry concentration; see Figure 5. The 'dilute' phase gas hold-up is found to depend on the superficial gas velocity through the dilute phase, $U - U_{df}$ and is practically independent of the slurry concentration (see Figure 6). This is a remarkable and useful result for scale up purposes as the large bubbles dictate gas phase conversion in slurry reactors operating in the heterogeneous regime[18]. The decrease in total gas voidage is almost entirely to be attributed to the decrease in the gas voidage of the 'dense' phase; see Figure 7. The physical rationalization of this observation is that the presence of solid particles tends to enhance the coalescence of 'small' bubbles while having no effect on the fast-rising 'large' bubbles. The void fraction of gas in the 'dense phase', ε_{df}, is found to be practically independent of the superficial gas velocity in the heterogeneous flow regime. The dense phase gas void fraction ε_{df}, was also found to correspond reasonably closely with the gas hold-up at which transition occurs from the homogeneous to the heterogeneous flow regime, ε_{trans}.

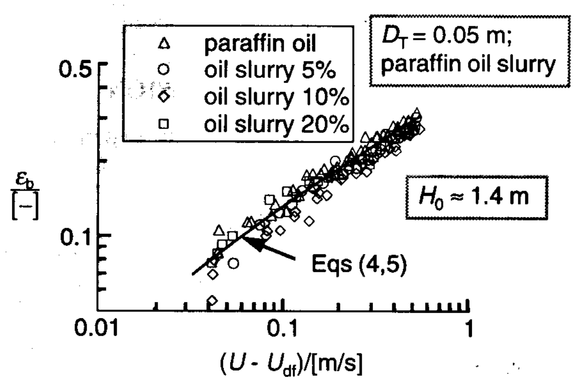

Figure 6. Influence of increasing particles concentration on gas voidage of 'dilute' phase.

MODEL FOR DILUTE PHASE GAS HOLD-UP

In a companion study on scale-up of gas-solid fluid beds and gas-liquid bubble columns, Ellenberger and Krishna[24] extended the bubble growth model of Darton *et al.*[25], developed for gas-solid fluid beds, to gas-liquid bubble columns. In this model the large bubbles are postulated to form as a result of coalescence of small bubbles following the picture in Figure 8. The coalescence process is limited to an equilibration height h^* above the distributor where the large bubbles reach their equilibrium size. The gas hold-up

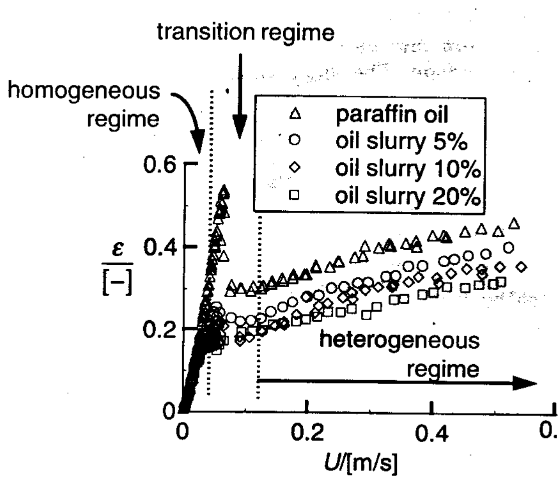

Figure 5. Influence of increasing particles concentration on total gas voidage.

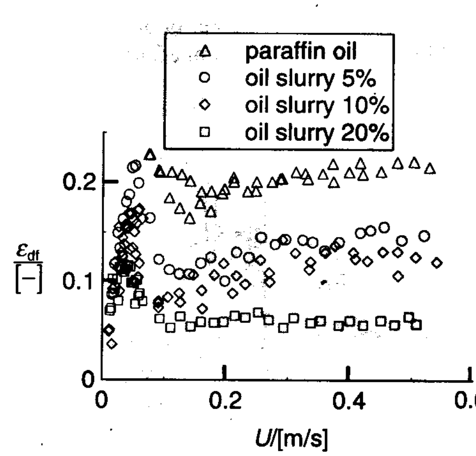

Figure 7. Influence of increasing particles concentration on gas voidage of 'dense' phase.

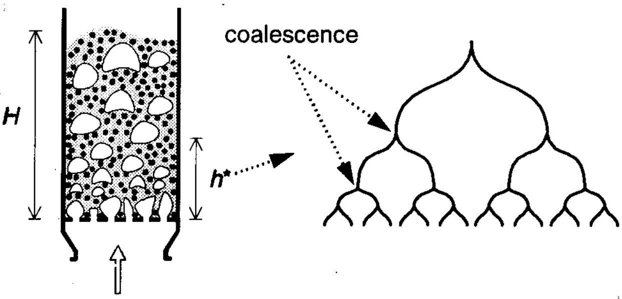

Figure 8. Coalescence model for 'dilute' phase gas. After Darton *et al.*[25].

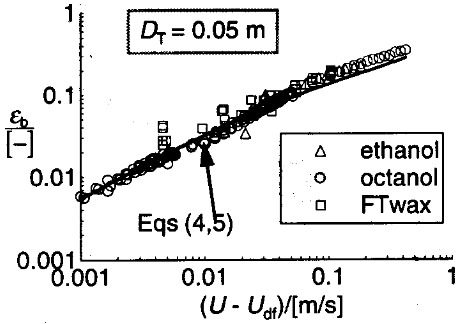

Figure 9. Dilute phase gas hold-up for various liquids in 0.05 m diameter column. Ethanol and octanol data are from present work. Fischer-Tropsch wax data from Daly *et al.*[23].

of the 'dilute' phase for a dispersion height H is

$$\varepsilon_b = \frac{1}{H}\int_0^{h*}\frac{(U-U_{df})}{V_b}\mathrm{d}h + \frac{1}{H}\int_{h*}^{H}\frac{(U-U_{df})}{V_b}\mathrm{d}h$$

The rise velocity of the 'large' bubbles, V_b is given by the relation $V_b = \phi_0 D_T^n\sqrt{gd_b}$, taking account of the influence of the column diameter on the rise velocity. The bubble diameter in the growth zone $0 - h^*$ is given by the Darton *et al.*[25] model to be $d_b = \alpha_1(U-U_{df})^{2/5}(h+h_0)^{4/5}g^{-1/5}$. Analytic integration gives the following expression:

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

Using extensive data for columns of 0.10, 0.19 and 0.38 m diameter, Ellenberger and Krishna[24] obtained the following values for the model parameters for paraffin oil

$$\alpha_1 = 1;\ h^* = 0.73(U-U_{df});\ \phi_0 = 1.95;\ n = 1/6 \tag{5}$$

The continuous line in Figure 6 has be drawn using equations (4) and (5), taking $D_T = 0.05$ m. The good agreement between the model predictions and the experimental data reinforces the validity of the model of Ellenberger and Krishna[24] to describe dilute phase hold-up in slurry systems as well.

INFLUENCE OF LIQUID PROPERTIES AND COLUMN DIAMETER ON DILUTE PHASE GAS HOLDUP

The independence of the dilute phase gas hold-up on slurry concentration raises the question whether the liquid phase properties have any influence at all on ε_b. Measurements were therefore made with ethanol and octanol in the 50 mm diameter column. These results are shown in Figure 9 along with the predictions of equations (4) and (5), *derived for paraffin oil.* The total gas voidage ε for ethanol and octanol are significantly different (cf. Figure 10); this difference is to be attributed to the differences in the gas voidage in the dense phase ε_{df}. Also plotted in Figure 9 is the large bubble data for Fischer-Tropsch waxes measured by Bukur *et al.*[3,4] and Daly *et al.*[23] obtained in a 50 mm diameter column at temperatures ranging from 200–265°C. It is remarkable that equations (4) and (5) also apply to such a wide range of liquid properties.

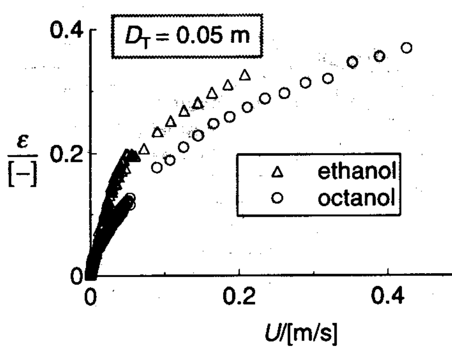

Figure 10. Total gas hold-up for ethanol and octanol in 0.05 m diameter column. Data from present work.

It can also be seen from equation (4) the dilute phase hold-up is a function of the column diameter. To test the validity of the model parameters in equation (5) we also carried out experiments in a 0.174 m diameter column fitted with a bronze sintered plate (pore size 50 μm). Figure 11 shows the dilute phase gas holdup ε_b for aqueous solutions of NaOH. The presence of electrolytes has no significant effect on e_b while the total gas voidage is affected in a pronounced manner; cf. Figures 11 and 12. Equations (4) and (5) are able to adequately predict the values of ε_b for electrolyte solutions. Further, the diameter effect is apparently also correctly reflected by equations (4) and (5).

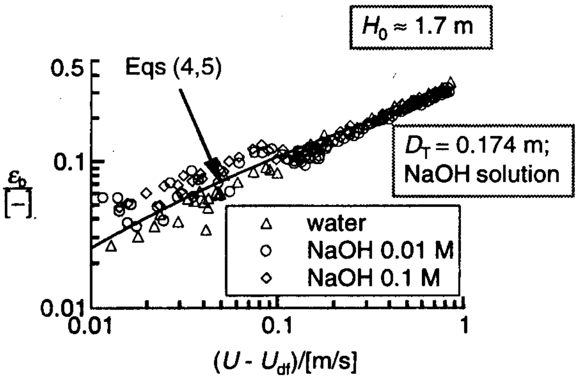

Figure 11. Dilute phase gas hold-up for aqueous NaOH in 0.174 m diameter column. Data from present work.

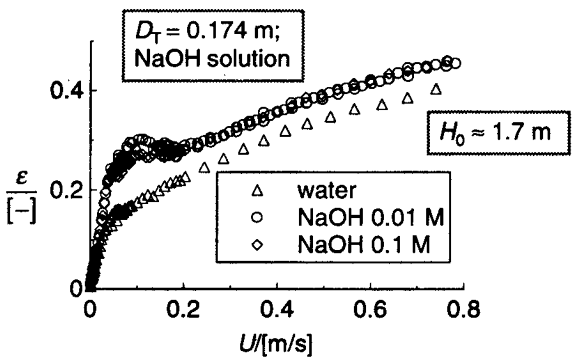

Figure 12. Total gas hold-up for aqueous NaOH in 0.174 m diameter column. Data from present work.

CONCLUSIONS

The dilute phase hold-up ε_b is practically independent of liquid properties and slurry concentration but does depend on the column diameter D_T and the dispersion height H. Equations (4) and (5) provide a practically usable model for prediction of the dilute phase hold-up. Since conversions in a bubble column slurry reactor are dictated by the fast rising 'dilute' phase[18], the present study provides a simple and useful scale up tool.

In the heterogeneous flow regime, the dense phase gas voidage ε_{df} is practically independent of the superficial gas velocity and decreases significantly with increasing particles concentration. The prediction of this parameter as a function of liquid properties and slurry concentration is an important aspect which deserves further detailed attention.

NOTATION

d_b	bubble diameter of dilute phase, m
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
H	height of expanded bed, m
H_0	height of ungassed bed, m
H_1	height of dispersion after escape of dilute phase, m
n	power in the rise velocity correlation, see equation (5)
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}
V_b	rise velocity of the dilute phase, m s^{-1}

Greek letters

α_1	constant, see equations (4) and (5)
ε	total gas voidage of G-S or G-L system
ε_b	gas hold-up of 'dilute' phase
ε_{df}	hold-up of gas in 'dense' phase
ε_{trans}	gas hold-up at the regime transition point
μ_L	liquid viscosity, Pa s
ρ_L	liquid density, kg m^{-3}
σ	surface tension of liquid phase, N m^{-1}
ϕ_0	constant, see equations (4) and (5)

REFERENCES

1. Henkel, K.-D., 1992, Reactor types and their industrial applications, Chapter in *Ullmann's Encyclopedia of Industrial Chemistry*, Volume B4, Principles of Chemical Reaction Engineering and Plant Design Editors: B. Elvers, S. Hawkins and G. Shulz, 5th Edition, (VCH Verlagsgesellschaft mbH, Weinheim) pp. 87–120.
2. Eisenberg, B., Ansell, L. L., Fiato, R. A. and Bauman, R. F., 1994, Advanced gas conversion technology for remote natural gas utilization, *73rd Annual GPA convention, March 7–9, 1994, New Orleans, Louisiana.*
3. Bukur, D. B., Patel, S. A. and Matheo, R., 1987, Hydrodynamics studies in Fischer-Tropsch derived waxes in a bubble column, *Chem Eng Commun*, 60: 63–78.
4. Bukur, D. B., Patel, S. A. and Daly, J. G., 1992, Gas holdup and solids dispersion in a three phase slurry bubble column, *AIChEJ*, 36: 1731–1735.
5. Deckwer, W.-D., Louisi, Y., Zaidi, A. and Ralek, M., 1980, Hydrodynamic properties of the Fischer-Tropsch slurry process, *Ind Eng Chem Proc Des Dev*, 19: 699–708.
6. Deckwer, W.-D. and Schumpe, A., 1993, Improved tools for bubble column reactor design and scale-up, *Chem Eng Sci*, 48: 889–911.
7. Deckwer, W.-D., 1992, *Bubble Column Reactors*, (John Wiley, New York).
8. Fukuma, M., Muroyama, K. and Yasunishi, A., 1987, Properties of bubble swarm in a slurry bubble column, *J Chem Eng Japan*, 20: 28–33.
9. Kara, S., Kelkar, B. G., Shah, Y. T. and Carr, N. L., 1982, Hydrodynamics and axial mixing in a three-phase bubble column, *Ind Eng Chem Proc Des Dev*, 21: 584–594.
10. Kelkar, B. G., Shah, Y. T. and Carr, N. L., 1984, Hydrodynamics and axial mixing in a three-phase bubble column. Effects of slurry properties, *Ind Eng Chem Proc Des Dev*, 23: 308–313.
11. Koide, K., Takazawa, A., Komura, M. and Matsunaga, H., 1984, Gas hold-up and volumetric liquid-phase mass transfer coefficient in solid-suspended bubble columns, *J Chem Eng Japan*, 17: 459–466.
12. O'Dowd, W., Smith, D. N., Ruether, J. A. and Saxena, S. C., 1987, Gas and solids behaviour in a baffled and unbaffled slurry bubble column, *AIChEJ*, 33: 1959–1970.
13. Saxena, S. C., Rao, N. S. and Thimmapuram, P. R., 1992, Gas phase holdup in slurry bubble columns for two- and three-phase systems, *Chem Eng Jl*, 49: 151–159.
14. Saxena, S. C. and Rao, N. S., 1993, Estimation of gas holdup in slurry bubble with internals: nitrogen-Therminol-magnetite system, *Powder Technology*, 75: 153–158.
15. Saxena, S. C., Rao, N. S. and Saxena, A. C., 1992, Heat transfer and gas phase holdup studies in a bubble column: air-water-sand system, *Canad J Chem Eng*, 70: 33–41.
16. Schumpe, A. and Grund, G., 1986, The gas disengagement technique for studying gas holdup structure in bubble columns. *Canad J Chem Eng*, 64: 891–896.
17. Schumpe, A., Saxena, A. K. and Fang, L. K., 1987, Gas/liquid mass transfer in a slurry bubble column, *Chem Eng Sci*, 42: 1787–1796.
18. Shah, Y. T., Joseph, S., Smith, D. N. and Ruether, J. A., 1985, Two-bubble class model for churn-turbulent bubble column reactor, *Ind Eng Chem Proc Des Dev*, 24: 1096–1104.
19. Shetty, S. A., Kantak, M. V. and Kelkar, M. G., 1992, Gas-phase backmixing in bubble column reactors, *AIChEJ*, 38: 1013–11026.
20. Van Deemter, J. J., 1961, Mixing and contacting in gas-solid fluidized beds, *Chem Eng Sci*, 13: 143–154.
21. May, W. G., 1959, Fluidized bed reactor studies, *Chem Eng Prog*, 55: 49–56.

22. Vermeer, D. J. and Krishna, R., 1981, Hydrodynamics and mass transfer in bubble columns operating in the churn-turbulent regime, *Ind Eng Chem Proc Des Dev*, 20: 475–482.
23. 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 Eng Sci*, 47: 3647–3654.
24. Ellenberger, J. and Krishna, R., 1994, A unified approach to the scale-up of gas-solid fluidized bed and gas-liquid bubble column reactors, *Chem Eng Sci*, (in press).
25. Darton, R. C., LaNauze, R. D., Davidson, J. F. and Harrison, D., 1977, Bubble growth due to coalescence in fluidized beds, *Trans Chem E*, 55: 274–280.

ACKNOWLEDGEMENTS

The experimental work reported in this paper was performed with the assistance of D. Barkey and R. Floore. Valuable comments and suggestions were received from J. Ellenberger.

ADDRESS

Correspondence concerning this paper should be addressed to Professor R. Krishna, Department of Chemical Engineering, University of Amsterdam, Nieuwe Achtergracht 166, 1018 WV Amsterdam, The Netherlands.