Large Bubble Sizes and Rise Velocities in a Bubble Column Slurry Reactor

By *C. O. Vandu*, *K. Koop,* and *R. Krishna**

The results are reported of an experimental study of the gas holdup, ε_G, large bubble diameter, d_{Lb}, and large bubble rise velocity, V_{Lb}, in a 0.1 m wide, 0.02 m deep and 0.95 m high rectangular slurry bubble column operated at ambient temperature and pressure conditions. The superficial gas velocity U was varied in the range of 0–0.2 m/s, spanning both the homogeneous and heterogeneous flow regimes. Air was used as the gas phase. The liquid phase used was C_9-C_{11} paraffin oil containing varying volume fractions (ε_S = 0, 0.05, 0.10, 0.15, 0.20 and 0.25) of porous catalyst (alumina catalyst support, 10 % < 10 μm; 50 % < 16 μm; 90 % < 39 μm). With increasing slurry concentrations, ε_G is significantly reduced due to enhanced bubble coalescence and for high slurry concentrations the "small" bubbles are significantly reduced in number. By the use of video imaging techniques, it was shown that the large bubble diameter is practically independent of the gas velocity for $\varepsilon_S > 0.05$ and $U > 0.1$ m/s. The measured large bubble rise velocity V_{Lb} agrees with the predictions of a modified Davis-Taylor relationship.

1 Introduction

There is currently a great deal of academic and industrial interest in the Fischer-Tropsch (FT) synthesis in the context of the conversion of remote natural gas to liquid transportation fuels. It is now widely accepted that the bubble column slurry reactor is the best choice of reactor type for large-scale plants with capacities of the order of 40,000 bbl/day of liquid hydrocarbon product [1–5]. The superficial gas velocity U in the FT bubble column reactor is in the range of 0.1–0.3 m/s depending on the catalyst activity and the catalyst concentration in the slurry phase [6]. For high reactor productivities, the highest slurry concentrations consistent with catalyst handling should be used. In practice, the volume fraction of catalyst in the slurry phase, ε_S, is of the order of 0.15–0.3 [6, 7]. At these high slurry concentrations the gas dispersion consists predominantly of fast-rising "large" bubbles [8]. The economic success of the FT process largely depends on the ability to achieve deep syngas conversions, say exceeding 95 %. Reliable design of the reactor to achieve such high conversion levels, requires reasonably accurate information on gas holdup, ε_G, and the volumetric mass transfer coefficient, k_La.

For the determination of ε_G it is important to have a good estimate of the rise velocity, V_{Lb}, of the "large" bubbles. For the determination of k_La it is important to have a good estimate of the size of the "large" bubbles. None of the published correlations for ε_G and k_La for [9–12] can be applied with confidence for such estimations. The major objective of this study is to gain insight into the hydrodynamics of bubble columns operating at gas velocities and slurry concentrations relevant for the FT synthesis. In particular, the emphasis is on the size and rise velocity of "large" bubbles and video imaging techniques are used to obtain the desired data by means of a rectangular bubble column. A predominantly C_9-C_{11} n-paraffin oil fraction and fine alumina catalyst carrier particles are utilized as the liquid and solid phases, respectively. The effects of slurry concentration and superficial gas velocity on gas holdup, bubble diameter and bubble rise velocity were investigated for superficial gas velocities ranging to 0.2 m/s.

2 Experimental

The experiments were conducted in a rectangular bubble column of 0.1 m width and 0.02 m depth, shown schematically in Fig. 1. The bubble column was fabricated from glass and had a perforated brass plate gas distributor with holes of 0.5 mm arranged on a triangular pitch of 7 mm, yielding a total of 43 holes. Gas was fed into the column with a precalibrated *Sho-Rate Brooks* rotameter and evaporating liquid was safely vented from the top of the column.

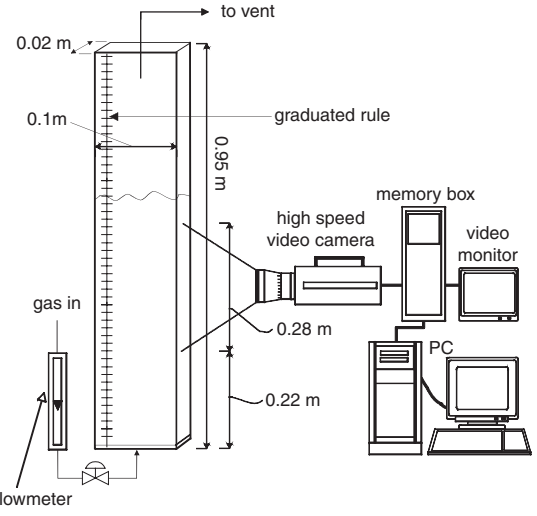

Figure 1. Rectangular slurry bubble column experimental setup. Further details are available on the authors' website [13].

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An image recording and analysis system was used to obtain and process data from the experimental runs. The image recording system consisted of a *Photron Fastcam-ultima 40 K* high-speed video camera connected to a 512 MB-memory box and a video monitor display. The high-speed camera has the capability of capturing video movies at rates of between 30 and 4500 frames per second (fps). It was positioned in front of the rectangular bubble column such that it captured a 0.28 m wide and 0.28 m high window. The base of this capture window was 0.22 m from the bottom of the column (see Fig. 1), high enough to ensure that bubble flow was satisfactorily developed and uninfluenced by gas distributor effects.

Lighting was provided by a single *Dedotec dedocool* 250 W Halogen Photo Optic Lamp. This lamp had the unique property of providing sufficient illumination without increasing the ambient temperature. Video movies captured by the high-speed camera were instantaneously stored in the memory box. The video monitor showed in real time what was viewed through the high-speed video camera. Data from the memory box were transferred to a PC for analysis. Further details on the setup are available elsewhere [13].

Air was employed as the gas phase in all experiments carried out with a predominantly C_9-C_{11} n-paraffin oil cut used as the liquid phase. Sasol PURALOX ScCa 5/170, an alumina-based catalyst particle carrier was employed as the solid phase with the slurry concentration, ε_S, varied in the range of 0, 0.05, 0.1, 0.15, 0.2 and 0.25. Note that slurry concentration is defined throughout this paper as the volume fraction of solids in gas-free slurry. The pore volume of the catalyst particles, which is liquid filled during experiments, was counted as being part of the solid phase. The properties of the paraffin oil and alumina particles utilized are given in Tabs. 1 and 2. The oil and catalyst properties correspond closely to those encountered in the FT reactor under reaction conditions. At the start of each experiment, the clear liquid or slurry height, H_0, was set at 0.5 m (for ε_S = 0 and 0.05 experiments) and 0.55 m (for ε_S = 0.1, 0.15, 0.2, 0.25 experiments).

Table 1. Properties of paraffin oil at 298 K.

Density	726 kg/m^3
Viscosity	0.85 mPa s
Surface tension	23.2 mN/m
Paraffin oil composition	≤ C_8 : 3.3 %; C_9 : 36.3 %; C_{10} : 34.5 %; C_{11} : 23.8 %; > C_{12} : 1.9 %

Table 2. Properties of catalyst carrier.

Al_2O_3 content	98 %
Skeletal density	3900 kg/m^3
Specific surface area	192 m^2/g
Particle porosity	70 %
Particle size distribution	10 % < 10 μm; 50 % < 16 μm; 90 % < 39 μm

Gas holdup was determined by visual measurements. For each run, the gas flow rate was adjusted with sufficient time given for steady state to be reached in the column after which the increase in dispersion height was recorded. The total gas holdup, ε_G, is defined as[1)]

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

where H_0 is the ungassed column height and H is the column dispersion height due to the presence of gas bubbles.

High-speed movies were recorded for superficial gas velocities U in the range of 0–0.2 m/s. The movies were recorded at 125 fps for a span of 8 seconds. Normal playback was at a rate of 30 fps (a set standard on Apple QuickTime and Microsoft Windows Media Player was used) implying that the movies were 4.17 times slower than real time when replayed. Playback speed was made slower when necessary by altering settings on the media players used. The movies were analyzed to determine the large bubble diameter and large bubble rise velocity. Samples of the video recordings can be viewed on the author's website [13].

3 Results and Discussion

Fig. 2 shows the measured gas holdup for varying slurry concentrations. Addition of catalyst particles tends to reduce the gas holdup, ε_G, to a significant extent, consistent with earlier work in cylindrical bubble columns [8,14]. The reduction in gas holdup with increasing ε_S is due to a decrease in the small bubble population. As illustration consider operation at U = 0.2 m/s; video images of the column for various slurry concentrations are shown in Fig. 3. For ε_S = 0, i.e., pure paraffin oil, a considerable fraction of the gas is present in the form of bubbles smaller than about 3 mm. Visual examination of the video images for ε_S > 0.1 showed also that

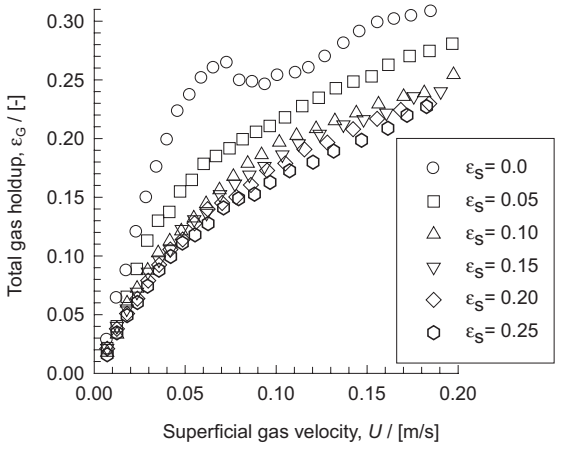

Figure 2. Influence of superficial gas velocity, U, on total gas holdup, ε_G, for varying slurry concentration, ε_S.

1) List of symbols at the end of the paper.

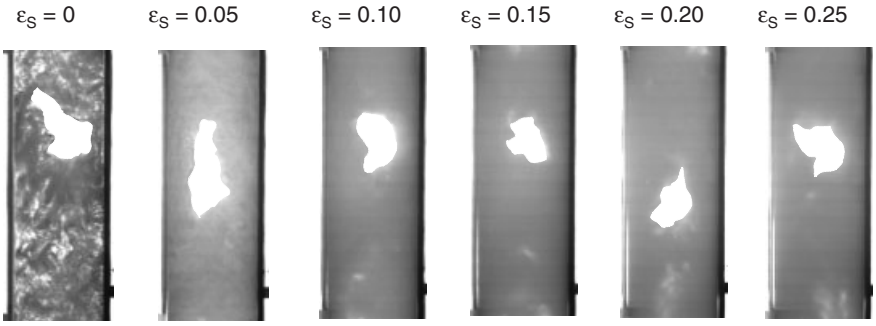

Figure 3. Video images obtained from a rectangular column operating at U = 0.2 m/s at various slurry concentrations showing the traced large bubbles after appropriate thresholding.

the dispersion consists predominantly of large bubbles for the whole range of values of U.

The transition gas velocity, U_{trans}, i.e., the gas velocity at which the first "large" bubble makes its appearance, was determined by examining the video images, frame by frame, of the column operation at different gas velocities. The data on U_{trans} as a function of ε_S is shown in Fig. 4a). It is noted that with increasing slurry concentration the "window" of operation of the column in the homogeneous regime becomes progressively narrower. The corresponding gas holdup at the regime transition point, ε_{trans}, is obtained from the gas holdup data and the values are shown in Fig. 4b). For gas velocities $U < U_{trans}$, the slope of the ε-U curve yields the rise velocity of the small bubbles, V_{Sb}, and the data are shown in Fig. 4c). It is noted that the small bubble rise velocity increases with increasing slurry concentration, an observation that is in line with previous work with a different oil-catalyst combination [8]. The reason for the increase in V_{Sb} with increasing ε_S is that the small bubble sizes are increased because of enhanced coalescence.

Attention now focuses on the "large" bubbles. For each run, a total of ten large bubbles were selected at random, from the hundreds of frames available from the movie recordings, and these had their areas computed. This was done by first making bitmap images of the particular movie frames containing the bubbles to be analyzed. Each image was then loaded into MATLAB. Utilizing certain MATLAB image processing functions which were coded in a program, the bubble image was traced out with a mouse and the area of the resulting polygon determined. Fig. 3 shows typical images after the large bubbles are identified and traced. From the projection areas, A_{Lb}, thus determined, the large bubble diameter, d_{Lb}, was calculated from:

$$d_{Lb} = \sqrt{\frac{4A_{Lb}}{\pi}} \tag{2}$$

The bubble diameters thus calculated are shown in Figs. 5a) to e) as a function of U for various slurry concentrations. The error bars represent the standard deviations of the bubble size determinations. The bubble sizes are considered as obtained in a rectangular bubble column. Fig. 5 gives a good indication of the actual bubble size in a cylindrical column. It is noted that for $\varepsilon_S > 0.05$ and $U > 0.1$ m/s, the d_{Lb} values show only a weak dependence on U. It is also noted that the bubble sizes for $\varepsilon_S > 0.05$ are practically independent of slurry concentrations. The results shown in Fig. 5 provide an explanation for the constancy of $(k_L a/\varepsilon)$ with slurry concentration and gas velocity in the experimental data reported by Vandu and Krishna [14]. The experimentally determined d_{Lb} values are also in reasonable agreement with the empirical correlation of Krishna *et al.* [15], developed for highly viscous Tellus oil (μ_L = 75 mPa s), indicated by the continuous line in Figs. 5a) to e):

$$d_{Lb} = 0.069\ \left(U - U_{trans}\right)^{0.376} \tag{3}$$

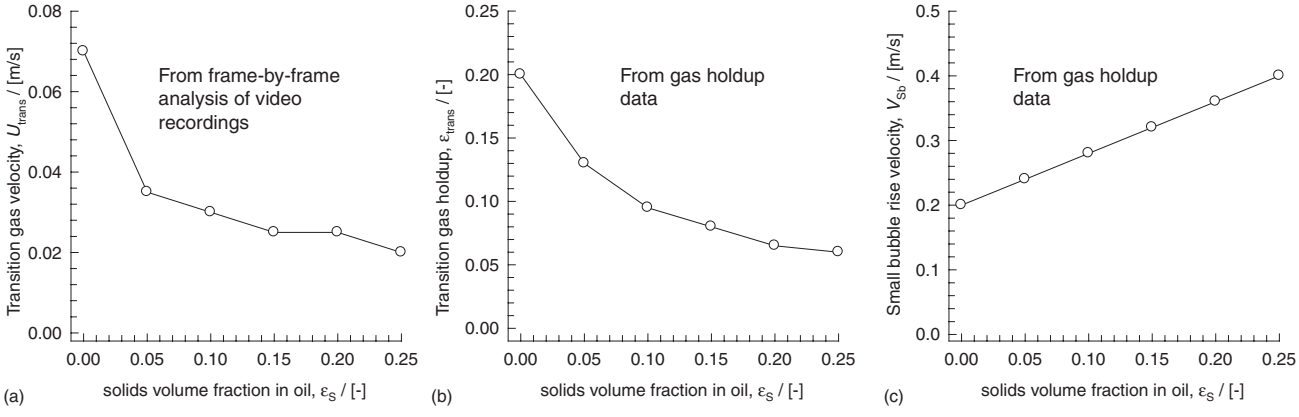

Figure 4. Influence of slurry concentration, ε_S, on (a) gas velocity at regime transition, U_{trans}, and (b) gas holdup at regime transition, ε_{trans}, and (c) rise velocity of the "small" bubble population, V_{Sb}.

The equivalence in the hydrodynamics of bubble columns operating with Tellus oil and concentrated paraffin oil slurries has been demonstrated in previous work [16].

The large bubble rise velocity, V_{Lb}, was determined from the high-speed movies by registering the time required for a large bubble to rise the 0.28 m height of the projection window. Each V_{Lb} reported is the average of five values taken, and the results are plotted in Fig. 5f) as a function of U for various slurry concentrations. The large bubble rise velocities are in excellent agreement with the extended Davies-Taylor relationship:

$$V_{Lb} = 0.71\sqrt{gd_{Lb}}(SF)(AF) \tag{4}$$

suggested by Krishna *et al.* [15], wherein the scale correction factor SF:

$$\begin{aligned} &SF = 1, \text{ for } d_{Lb}/D_T < 0.125; \\ &SF = 1.13\ \exp(-d_{Lb}/D_T) \text{ for } 0.125 < d_b/D_T < 0.6; \\ &SF = 0.496\sqrt{D_T/d_{Lb}} \text{ for } d_b/D_T > 0.6 \end{aligned} \tag{5}$$

and the acceleration factor AF (also for Tellus oil):

$$AF = 2.25 + 4.09(U - U_{trrans}) \tag{6}$$

This acceleration is due to wake interactions and this factor increases as the distance between the large bubbles decreases.

From the knowledge of the large bubble sizes and rise velocity it is now possible to estimate the total gas holdup, ε, for a bubble column slurry reactor. In the homogeneous regime

$$\varepsilon = \frac{U}{V_{Sb}}; \quad U < U_{trans} \tag{7}$$

whereas in the heterogeneous flow regime

$$\varepsilon = \frac{(U - U_{trans})}{V_{Lb}} + \varepsilon_{trans}\left[1 - \frac{(U - U_{trans})}{V_{Lb}}\right]; \quad U > U_{trans} \tag{8}$$

The calculations of the total gas holdup by using Eqs. (7) and (8) are shown in Fig. 6 where U_{trans}, ε_{trans}, and V_{Sb} are taken from the data shown in Fig. 4. It is noted that the total gas holdup for $\varepsilon_S = 0.2$ and $\varepsilon_S = 0.25$ are practically the same. The total gas holdup predictions are also in good agreement with the experimental data presented in Fig. 2.

4 Conclusions

The sizes and rise velocity of "large" bubbles have been measured in a rectangular bubble column operating with concentrated slurries at high superficial gas velocities. The following major conclusions can be drawn from this work.

– The gas holdup is significantly reduced when the concentration of catalyst is increased. This reduction in gas hold-

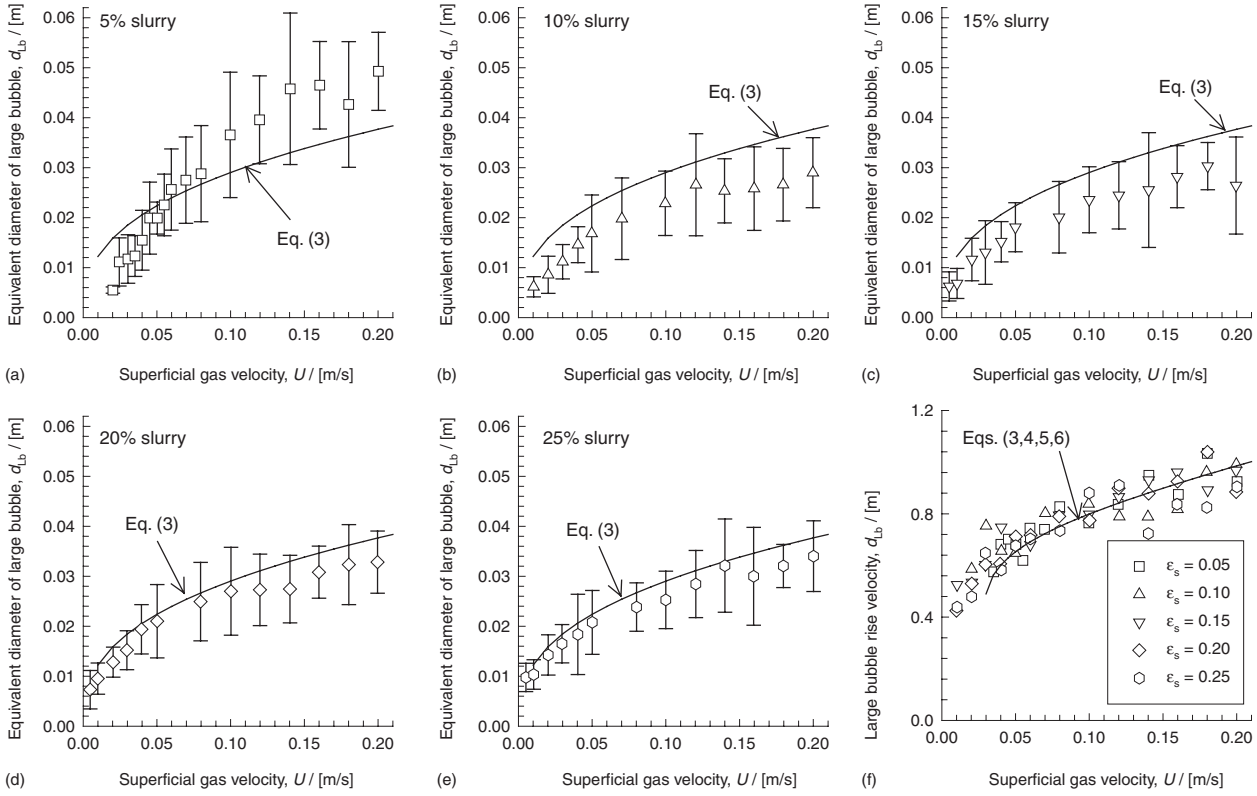

Figure 5. Large bubble diameter, d_{Lb}, for slurry concentration ε_S = (a) 0.05, (b) 0.1, (c) 0,15, (d) 0.2 and (e) 0.25; (f) large bubble rise velocity, V_{Lb}, as a function of U for various slurry concentrations, ε_S.

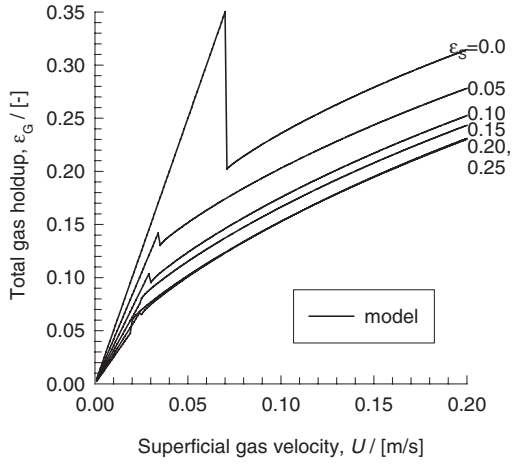

Figure 6. The total gas holdup for a bubble column slurry reactor as a function of the superficial gas velocity and various slurry concentrations. The calculations use Eqs. (7) and (8).

up is primarily due to the reduction in the holdup of the small bubble population.

- The window of operation of a slurry bubble column is made considerably narrower when the slurry concentration is increased.
- Video imaging shows that the large bubble diameter, d_{Lb}, is practically independent of the gas velocity, U, for $\varepsilon_S > 0.05$ and $U > 0.1$ m/s. Eq. (3) provides a reasonably good estimate of the bubble size. The information on the large bubble sizes provides clues to the interpretation of published k_La for slurry bubble columns [14].
- The rise velocity of large bubbles in slurries, V_{Lb}, can be accurately predicted with the extended Davis-Taylor relationship (4) using the appropriate scale correction factor (5) and the acceleration factor (6) suggested based on earlier work with Tellus oil.
- Eqs. (7) and (8) provide good estimates of the total gas holdup in slurry bubble columns.

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Symbols used

A_{Lb}	[m^2]	projected area of large bubble from video images
AF	[–]	acceleration factor, dimensionless
d_{Lb}	[m]	diameter of large bubble
D_T	[m]	column diameter
H	[m]	dispersion height in the column
H_0	[m]	height of ungassed column
SF	[–]	scale correction factor, dimensionless
U	[m/s]	superficial gas velocity
V_{Lb}	[m/s]	rise velocity of large bubbles
V_{Sb}	[m/s]	rise velocity of small bubbles

Greek symbols

ε_G	[–]	total gas holdup, dimensionless
ε_S	[–]	volume fraction of catalyst in the slurry, dimensionless
μ_L	[Pa s]	liquid viscosity

Subscripts

G	referring to gas phase
L	referring to liquid
Lb	referring to large bubble
trans	referring to the regime transition point
S	referring to porous solid (catalyst)
T	tower or column

References

[1] B. Eisenberg, R. A. Fiato, T. G. Kaufmann, R. F. Bauman, *Chemtech.* **1999**, *29*, 32.
[2] R. L. Espinoza, A. P. Steynberg, B. Jager, A. C. Vosloo, *Appl. Catal. A* **1999**, *186*, 13.
[3] B. H. Davis, *Catal. Today* **2002**, *71*, 249.
[4] S. T. Sie, R. Krishna, *Appl. Catal. A* **1999**, *186*, 55.
[5] R. Krishna, S. T. Sie, *Fuel Process. Technol.* **2000**, *64*, 73.
[6] C. Maretto, R. Krishna, *Catal. Today* **1999**, *52*, 279.
[7] C. Maretto, R. Krishna, *Catal. Today* **2001**, *66*, 241.
[8] R. Krishna, J. W. A. de Swart, J. Ellenberger, G. B. Martina, C. Maretto, *AIChE J.* **1997**, *43*, 311.
[9] K. Akita, F. Yoshida, *Ind. Eng. Chem. Process Des. Dev.* **1973**, *12*, 76.
[10] K. Akita, F. Yoshida, *Ind. Eng. Chem. Process Des. Dev.* **1974**, *13*, 84.
[11] J. R. Inga, B. I. Morsi, *Can. J. Chem. Eng.* **1997**, *75*, 872.
[12] J. R. Inga, B. I. Morsi, *Ind. Eng. Chem. Res.* **1999**, *38*, 928.
[13] C. O. Vandu, K. Koop, J. Ellenberger, R. Krishna, Hydrodynamic Studies in a Rectangular Slurry Bubble Column, http://ct-cr4.chem.uva.nl/RectBubbleColumn/, **2004**.
[14] C. O. Vandu, R. Krishna, *Chem. Eng. Technol.* **2003**, *26*, 779.
[15] R. Krishna, M. I. Urseanu, J. M. van Baten, J. Ellenberger, *Chem. Eng. Sci.* **1999**, *54*, 171.
[16] R. Krishna, M. I. Urseanu, J. W. A. de Swart, J. Ellenberger, *Can. J. Chem. Eng.* **2000**, *78*, 442.

