A structured catalytic bubble column reactor: hydrodynamics and mixing studies

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Abstract

This paper reports the results of a comprehensive experimental study of the hydrodynamics and mixing in two bubble column reactors of 0.1 and 0.24 m in diameter with KATAPAK-S$^{®}$ as packing material. Total gas hold up and axial dispersion coefficients were measured in the structured bubble columns and the values were compared with experimental results obtained in the same work with empty bubble columns. The results reveal that the gas hold up in structured bubble columns is practically the same as in empty bubble columns when compared at the same superficial gas velocity based on open area available for gas–liquid dispersion. The presence of the structured elements in the bubble column reactor reduces the liquid phase backmixing by one order of magnitude. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

A bubble column slurry reactor is often used in industrial practice, e.g., in the Fischer–Tropsch synthesis of heavy paraffins from syngas (CO + H$_2$) [1,2]. The bubble column slurry reactor has complex hydrodynamics is difficult to scale up and the separation of the catalyst particles from the product of liquid hydrocarbons is a difficult and expensive step. We are studying an alternative concept to the conventional slurry reactor: the structured catalytic bubble column reactor. A schematic drawing of this reactor concept is shown in Fig. 1.

Structured catalytic bubble columns are new, promising types of multiphase reactors. Their configuration lies basically between slurry reactors and trickle bed reactors. The reactor is made up of several structured catalytic sections. Each section consists of KATAPAK-S elements, supplied by Sulzer Chemtech. The structure consists of two pieces of rectangular crimped wire gauze sealed around the edges, thereby forming a pocket of the order of 10–50 mm wide between the two screens (see Fig. 1). The inclination of the corrugated sheets is $\alpha = 45°$. The catalytic particles, of say 1 mm in diameter, are enveloped within wire gauze envelopes. Conventionally, structured catalytic reactors have been operated in the gas-continuous regime with trickling liquid flow within the packing elements [3–9]. We believe that there is a distinct advantage of operation in the liquid-continuous regime, i.e., bubble column operation. There is no published information on operation in this regime. In this regime of operation, the gas flows preferentially in the open channels, between the wire gauze envelopes. The liquid flows down the structures both within the packed channels and in the open

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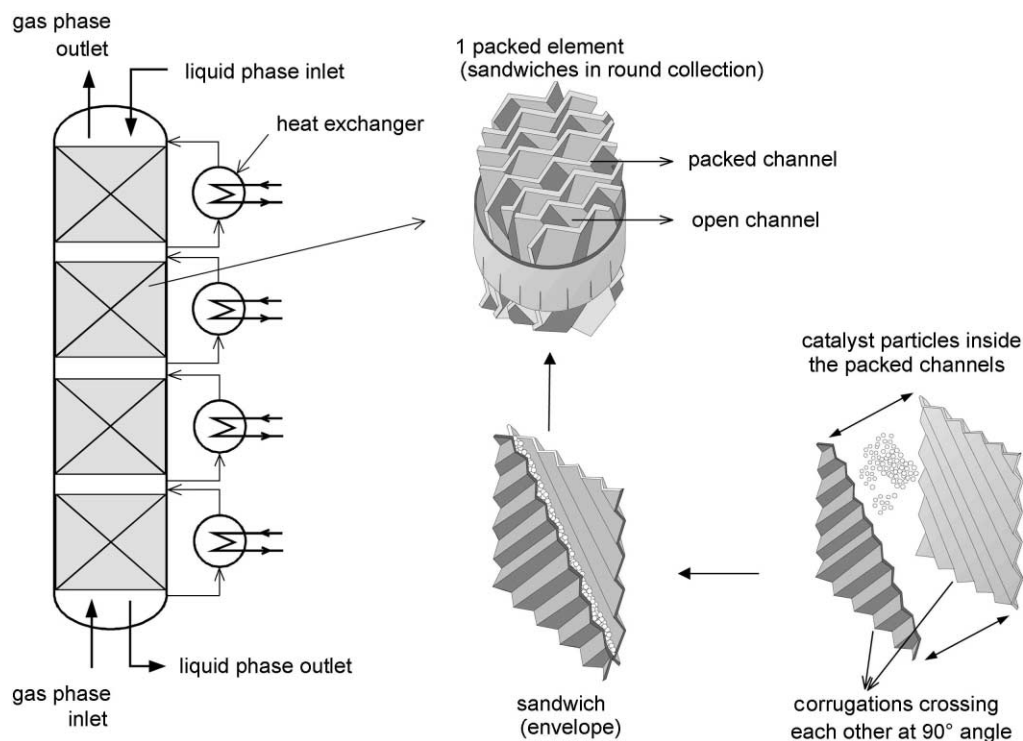*E-mail address:* krishna@its.chem.uva.nl (R. Krishna).

Fig. 1. The structured bubble column reactor concept (including details of a packed element).

channels. These catalyst "sandwiches" or "wafers" are bound together and installed as a monolith inside the column. Within the catalyst sandwiches, the liquid follows a criss-crossing flow path and the radial dispersion is about an order of magnitude higher than conventional packed beds. Furthermore, frequent criss-crossing leads to a significant improvement in mass transfer within the sandwich structures. The liquid can be made to flow co- or counter-current to the gas phase. The reaction enthalpy can be removed by external heat exchange as shown in Fig. 1.

The main advantages of this new reactor with respect to the conventional slurry column are: (1) no problems for separating catalyst from the liquid; (2) improved conversion and selectivity due to staging of the liquid phase, and (3) no scale up problems because hydrodynamics is dictated by the size of the open channels of the catalytic structure. The main advantages over trickle beds are: (1) lower pressure drop even with 1 mm size particles; (2) excellent

radial dispersion, and (3) possibility of counter-current operation without flooding.

Since there is almost no published literature on design of structured bubble column reactors, our main objective is to obtain design and scale up information on the hydrodynamics and mixing characteristics of such reactors by performing experiments.

2. Experimental

The studies were carried out in columns of 0.1 and 0.24 m in diameter; a typical set-up is shown in Fig. 2(a). The columns are equipped with perforated plates gas distributors. Gas was taken from the compressed air mains and introduced into the column from the base. The volumetric flow-rate was determined by a row of several calibrated Brooks flowmeters. The operating pressure was atmospheric for all the cases. The columns are divided into two
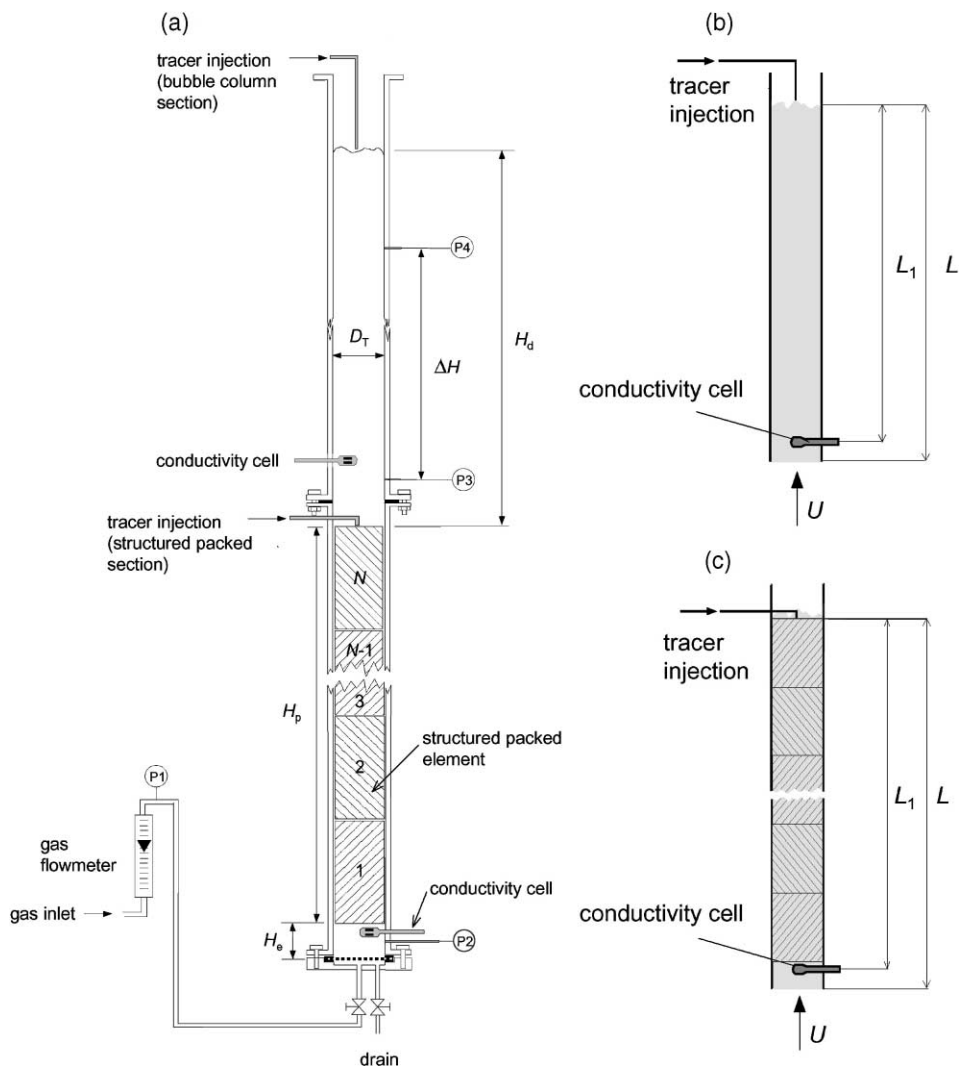
Fig. 2. (a) Typical experimental set-up. Injection point and conductivity cell position in the (b) bubble column section; (c) structured packed section.

segments: the bottom segment contains the structured packing; the upper part is kept empty, as is an ordinary bubble column. Complete details of the column configurations, including photos, are available on our web site: http://ct-cr4.chem.uva.nl/structuredbc/.

2.1. Gas hold up measurements

The gas hold up in the *bubble column section* was measured using a differential pressure sensor connected to the column. The continuously recorded output signal coming from the pressure sensor (Validyne DP15) is linearly dependent on the differential pressure $(P_4 - P_3)$ in the column. Prior to measurements, the pressure sensor has been carefully calibrated. The hold up measurements were performed for different superficial gas velocities measured at the bottom of the bubble column section (i.e., above the last packed element from the structured packed segment) (see also Fig. 2(a)). The dispersion height

in the bubble column, H_d, was kept constant for each given superficial gas velocity, by adjusting the volume of demineralised water into the column.

The gas hold up in the *structured packed bubble column section* was calculated indirectly, as a residual volume from the total reactor volume, after subtracting the solids (V_S) and the liquid part (V_L). From the total volume of the cylindrical tube, the fraction occupied by the solids was first subtracted. The solids consist of the wire mesh, together with the glass beads. The remaining volume, V_{OS}, represents the available open space in the column. The gas hold up in the structured packed section is therefore given by

$$\varepsilon_G = \frac{\text{volume of gas present in the packed structured column}}{\text{available "open space" in the packed structured column}} = \frac{V_{OS} - V_L}{V_{OS}} \quad (1)$$

The dispersion height was kept constant at the top level of the packed elements, $H_p + H_e$, for each experimented superficial gas velocity, measured at the bottom of the column, considering only the open cross-section area available for the gas flow (the open space area). The total volume of liquid was measured from the collected liquid mass. The height of the structured packed section (H_p) is 1.8 m for the 0.1 m column and 1.68 m for the 0.24 m diameter column.

2.2. RTD measurements

Liquid phase residence time distribution (RTD) measurements have been performed using a tracer of saturated NaCl solution. The solution was injected into the batch liquid phase as a pulse at the top of each reactor section. The transient salt concentration was monitored continuously by means of an immersing-type conductivity cell (Metrohm), which was placed at the bottom of each section of the column. The positions of the injection points and the conductivity cells in both sections of the column are shown in Fig. 2(b) and (c). Both injection tubes and conductivity cells were positioned in the centre axis of the column. Different volumes of tracer were used, depending on the amount of liquid present in the column for each gas velocity, in order to obtain the optimal signal. The measurements were carried out with constant dispersion height in both column sections. The measuring time was set in such a way

that the final (plateau) concentration in the column was reached.

2.3. RTD data analysis

Typical transient concentrations curves are shown in Fig. 3(a) and (c). Fig. 3(a) shows dimensionless salt tracer concentrations from the 0.1 m diameter column, for the bubble column section, operated at $U = 0.25$ m/s superficial gas velocity. Fig. 3(c) presents the signal recorded in the structured packed section of the 0.24 m diameter column, operated at $U = 0.21$ m/s superficial gas velocity. The details (b) and (d) represent the normalised concentration used for the fit, in each case. These signals were fitted using the analytic solution to the diffusion equation with the appropriate boundary conditions [10,11]:

$$\frac{C_L}{C_0} = 1 + 2\sum_{n=1}^{\infty} \left[\left(\cos\left(\frac{n\pi}{L}\right) L_1 \right) \exp \right.$$
$$\left. \times \left(-\frac{n^2\pi^2}{L^2} D_{ax,L} t \right) \right] \quad (2)$$

where C_L is the concentration of the tracer in the liquid phase, C_0 the final concentration achieved when $t = \infty$, L_1 the distance from top to the measuring point and L is the total dispersion height in the section of the column. For a given superficial gas velocity, only one variable was adjusted, i.e., the axial dispersion coefficient, $D_{ax,L}$. The axial dispersion coefficient $D_{ax,L}$ was obtained by adjusting the experimental profiles with the solution of the model, Eq. (2). A number of $n = 20$ terms were found to be sufficient.

In the bubble column section case, from the entire signal recorded (see Fig. 3(a)) a first portion was analysed, considered to reveal the mixing only due to the bubble column section. The signal was further fitted with the model only for this portion. The final concentration, C_0, in the bubble column section is reached within the first 120 s (depending on the gas velocity this time can vary). This value was used for normalisation of the data. This portion of the fitted signal is shown in Fig. 3(b). The further
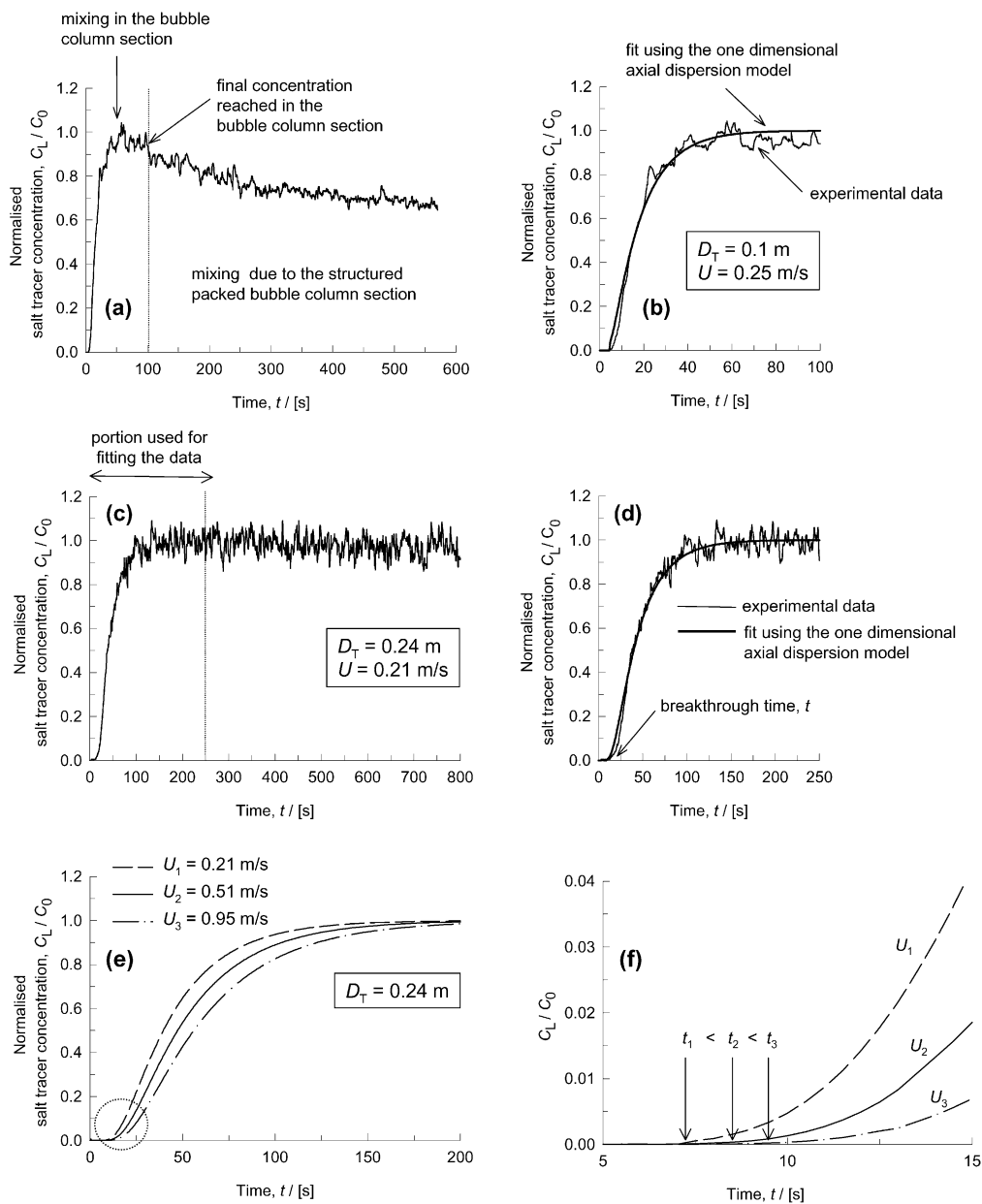
Fig. 3. Typical salt tracer concentration measured at the bottom of the (a) bubble column section in response to pulse tracer injection in the 0.1 m column; (c) structured packed section in response to pulse tracer injection in the 0.24 m column; details (b) and (d) represent the normalised concentration used for the fit. The smooth curve represents the fit to the curve using a diffusion model presented in [10]; details (e) and (f) represent the determination of breakthrough time for the structured packed column.

decrease in the concentration is supposed to be influenced by the mixing with the bottom section of the column.

From the signal recorded for the structured packed section, also a first portion was cut-off and used for fitting the data. In this way the procedure is considered to give a more accurate fit, since the rest of the data shows an almost constant value of the final concentration achieved in the reactor.

2.4. Characteristic liquid velocity determination

The transient concentration curves were also used to determine the breakthrough liquid velocity in the packed section of the column. The breakthrough time was determined from the experimental data, by reading the time necessary for the first salt tracer to arrive at the bottom of the column, where the conductivity cell was placed, as shown in Fig. 3(c). These values are used further to determine the breakthrough liquid velocity, $U_L = H_p/t$, and this velocity is a characteristic for the liquid circulation in the open channels. The liquid has a slower motion in the packed channels than in the open ones, also when operating at small superficial gas velocities (thus high liquid hold ups in the column).

As the superficial gas velocity increases, we would expect larger values for the breakthrough time. These results are shown in Fig. 3(e) and (f), where we compare the data obtained in the 0.24 m column, for three superficial gas velocities: $U = 0.21$, 0.51 and 0.95 m/s.

3. Results and discussion

3.1. Gas hold up

Comparison of the hold ups in the 0.1 and 0.24 m bubble columns (Fig. 4 (a)) confirm the findings of our previous work [12–14]. With increasing column diameter, the average hold up decreases. Fig. 4(b) compares the gas hold up in the structured packed sections of the 0.1 and 0.24 m column. We note that up to a superficial gas velocity of 0.6 m/s, the values are almost same for both columns. At superficial gas velocities larger than $U = 0.6$ m/s, a difference in the two trends arises, the larger hydraulic diameter of the packing giving a lower gas hold up. The mechanism of interaction between the gas bubbles in such packed geometry is different than the one in empty bubble columns. The bubble size is limited by the diameter of the open channels and the bubbles suffer continuous coalescence and break up, caused by the criss-crossing channels path. An explanation for the lower gas hold up in the 0.24 m column, compared with the 0.1 m one can be given by the larger size of the open channels. The hydraulic diameter of the open channels dictates the gas hold up. The values of the hydraulic diameter for the 0.1 and 0.24 m columns are 0.007 and 0.02 m, respectively.

If we compare the data in the structured packed section with that in the empty section of each column experimented, when compared at the same superficial gas velocity based on the open area available for the gas flow (Eq. (1)), the gas hold ups are practically the
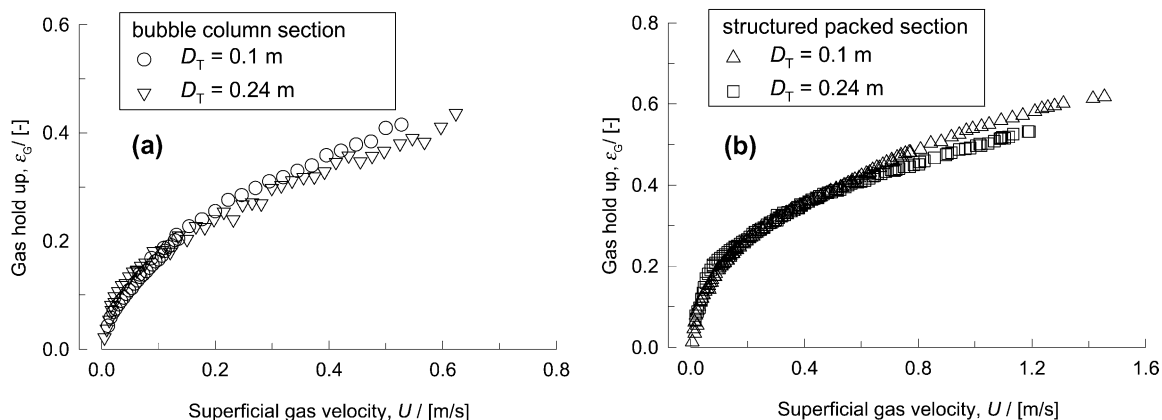

Fig. 4. Comparison of hold ups in the 0.1 and 0.24 m diameter columns: (a) bubble column section; (b) structured packed section.
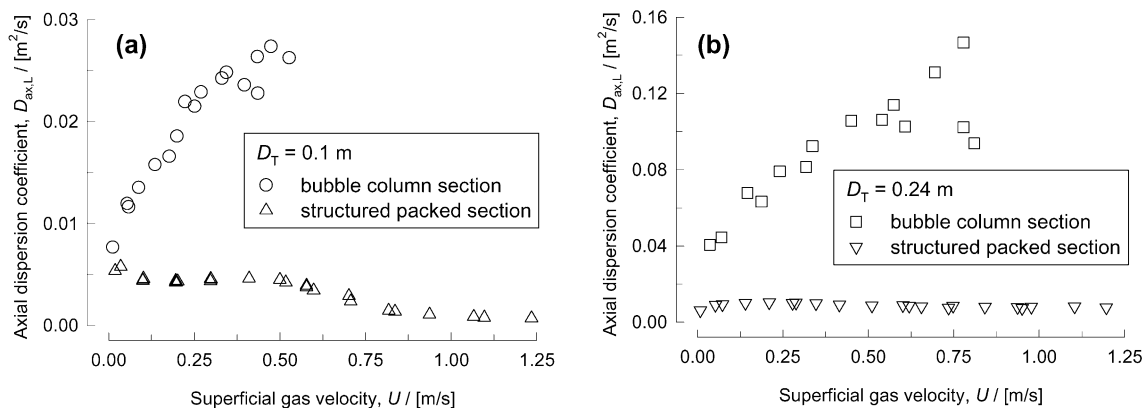
Fig. 5. Axial dispersion of the liquid phase; comparison between structured packed section and empty section for columns of: (a) $D_T = 0.1$ m and (b) $D_T = 0.24$ m.

same for both columns. There is no additional coalescence caused by the packing elements. We also note that the operation in the structured packed bubble column is possible up to high superficial gas velocities, of around 1.2–1.5 m/s. The model developed by Krishna et al. [12] was found to be applicable for estimating the gas hold up in both empty and structured bubble columns.

3.2. Mixing of the liquid phase

The presence of the structured elements in the reactor reduces the liquid phase backmixing by a factor of about one order of magnitude (see Fig. 5). The liquid phase in the structured bubble column is virtually in plug flow. When comparing the axial dispersion in the liquid phase data obtained in the bubble column sections of both reactors with previous research, we see that the data are well predicted using the model developed by Krishna et al. [11,13]:

$$D_{ax,L} = 0.31 V_L(0) D_T \tag{3}$$

The centre-line liquid velocity, $V_L(0)$, data used for Fig. 6(a) are experimental data (when data was available) or calculated values using the correlation of Riquarts [15]:

$$V_L(0) = 0.21\sqrt{D_T g}\left(\frac{U_G^3}{\nu_L g}\right)^{1/8} \tag{4}$$

In the packed column section axial mixing is the result of the combined effect of non-ideal flow situations (small density differences between phases, maldistribution of the two phases, turbulence, liquid exchange between the open and the packed channels, pulse flow in the open channels). The packed channels are assumed to be completely filled with liquid phase. At low gas velocities, the liquid phase in the packed channels is almost stagnant (due to the fact that the density of dispersion in the open channels is almost equal to that of the liquid phase in the packed channels) and the axial dispersion is dominated by the liquid phase in the open channels. We used as a measure for the characteristic liquid velocity in the open channels the breakthrough liquid velocity, U_L, calculated from the derived breakthrough times, as described above. The resulting axial dispersion coefficient is expected to be predicted by a model similar with Eq. (3) (the model developed for empty bubble columns by Krishna et al. [11,13]). The resulting equation is

$$D_{ax,L} = 0.081 U_L \tag{5}$$

The predictions of this model are shown in Fig. 6(b) together with the experimental data. The constant in Eq. (5) was chosen equal for both columns, although a slightly different values is obtained for each column.

With increasing the superficial gas velocity, the liquid phase fraction and the density of dispersion in the open channels decreases and the breakthrough liquid velocity in the packed channels will increase,
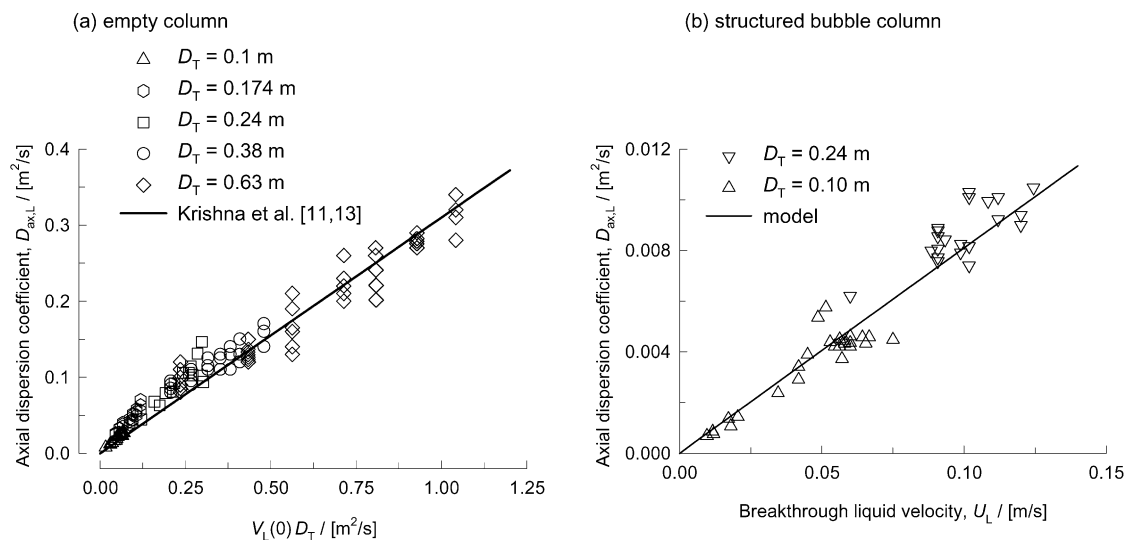
Fig. 6. (a) Axial dispersion coefficient of the liquid phase in bubble column as a function of $V_L(0)D_T$. Comparison between experiments obtained in this work and in our previous work and Krishna et al. [11,13] model predictions. (b) Comparison between the measured axial dispersion coefficient in the liquid phase, $D_{ax,L}$ and the predictions of Eq. (5).

resulting in a more pronounced exchange between open and packed channels. Therefore, at high gas velocities, the axial dispersion in the packed channels becomes more significant on the overall axial dispersion coefficient.

4. Conclusions

The main conclusions which may be drawn from the results presented in this work are:

1. The gas hold up in structured bubble columns is practically the same as in empty bubble columns.
2. Comparison of the gas hold up in the 0.1 and 0.24 m structured packed columns reveals a small difference starting with superficial gas velocities of about $U = 0.6$ m/s; larger column diameter gives lower gas hold up. The hydraulic diameter of the open channels dictates the gas hold up; the two columns have different hydraulic diameters.
3. The presence of the structured elements in the bubble column reactor reduces the liquid phase backmixing by a factor of about one order of magnitude.
4. The axial dispersion coefficient in structured bubble columns can be predicted using the correlation

given by Eq. (5) which shows that the axial dispersion coefficient is proportional to the breakthrough liquid velocity U_L. More detailed models will be required for a priori prediction of the breakthrough velocity.

5. Compared with a bubble column reactor, we may conclude that operation of a structured packed bubble column has the advantage of a reduced backmixing of the liquid phase, combined with a good catalyst contact efficiency. Moreover, in contrast to a bubble column, the superficial gas velocity in a structured packed bubble column, can be varied in a broad range, without significant change in the axial dispersion.

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