

IMPACT OF STRUCTURED PACKING ON BUBBLE COLUMN MASS TRANSFER CHARACTERISTICS

Part 1. Backmixing in the Liquid Phase[#]

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[#]This paper is dedicated to Professor Dr. Roman Modic

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Abstract

Axial liquid phase dispersion coefficient in a concurrent up-flow bubble column packed with Sulzer structured packing (SMV 16) was measured by means of the stationary method in a region between the homogeneous and heterogeneous hydrodynamic regimes. Column had a 0.14 cm ID and 1.885 m total packing length. Tap water was used as liquid phase and oxygen as gas phase. KCl was used as a tracer. For comparison experiments were also performed in the same column but without packing. The gas holdup was determined simultaneously. An increase in the measured E_L values with the gas velocity was observed in both packed and non-packed columns, while the liquid flow rate only slightly raised the axial dispersion coefficient when the column was packed. The presence of structured packing reduced the liquid axial dispersion coefficient for nearly 50 % at low gas velocities, whereas there was only 20 % reduction found at higher velocities.

Introduction

Bubble columns are widely used in industry as absorbers, fermentors and gas-liquid reactors. The gas phase continuously enters at the bottom of vertical column through a gas sparger and moves through the liquid phase in the form of bubbles. The liquid, which in this case acts as the continuous phase, may be either in concurrent or countercurrent flow with the gas. A solid reactant or catalyst particles may also be suspended in the liquid phase (bubble column slurry reactors). Depending on the nature of process the bubble columns can be found in many different modifications.¹ The use of different structured packing is a very common practice nowadays. As an absorber, the bubble column must provide an efficient contact between the flowing phases, which provides high interfacial area and therefore high mass transfer rates. When designing a system with simultaneous chemical reaction, some additional requirements must be

fulfilled. For example, in the case of a slow chemical reaction (the majority of oxidation, hydrogenation, etc.²), which calls for high contact times, high liquid holdup is desired for efficient process, but with fast high gas holdup with small bubbles ensures the needed interfacial area.

When studying the transport processes in bubble columns, the understanding of flow behavior is of utmost importance. The gas bubbles formed under required operating conditions are of non-uniform size and travel with different velocities, which is the main cause of non-ideal flow of gas. The rising gas bubbles carry the liquid upwards, which then returns in the circular flow. Back-mixing in the liquid phase causes a decrease of mass-transfer driving force and lowers the conversion of a reactant compared to the plug flow behavior. The best way to describe the departure from the ideal plug flow is to adopt the one-dimensional axial dispersion model (ADM)³, where the axial dispersion coefficient is understood as a lump parameter. It is, however, known that employing packing in bubble columns reduces channeling and promotes mixing in the radial direction, which in turn minimizes a negative effect of axial mixing in the liquid phase.

In the open literature many reliable correlations are available for evaluating the liquid phase dispersion coefficient in bubble column reactors. Unfortunately, data for modern packing (second generation) and structured packing in particular are very meager. This was recently revealed by Macias-Salinas and Fair⁴ who studied the axial mixing in column packed with modern packing (Raschig rings and Pall rings of second generation) and structured packing such as Sulzer BX and Flexipac 2. Their results with countercurrent flow confirmed previous observations, i.e. axial mixing in the gas phase increases with both gas and liquid flow rates, whereas the liquid phase axial dispersion coefficient is a decreasing function of liquid flow rate but is insensitive to gas flow rate up to the flooding point. In addition, they found the inclination of channels as well as channel size as the two of the most important factors in determining the extent of axial mixing. It is interesting to note that Belfares et al.⁵ criticized the existing correlations for bubble columns packed with spheres, cylinders, Raschig rings, and extrudates, in the sense that not all the variables affecting the dispersion coefficient (E_L) were accounted for. On the basis of a large database they developed a state-of-the-art correlation for

liquid axial dispersion coefficient by combining neural network modeling and dimensional analysis. Although 28 different packing materials were used in their study, no packing of the second generation, neither the structured packing was included in their work.

The aim of this work was to study the effect of structured packing (motionless or static mixers) on the liquid axial mixing in a concurrent bubble column with relatively high column height to diameter ratio. Gas and liquid flow rates were chosen in the region of operating conditions usually employed in wet oxidation processes. Since there is no literature data for structural packing of the type employed in this work, the results for structural packing were compared to those obtained at the same operating conditions in bubble column without packing. However, the extent of axial mixing was evaluated by means of correlations that are available for bubble columns. Therefore, a brief overview of the correlations for axial dispersion coefficient in bubble columns is presented below.

Axial dispersion in bubble columns-literature survey

Most of the experimental data on axial dispersion in bubble columns can be interpreted by the theoretical relation of Baird and Rice.⁶ They used the Kolmogoroff theory of isotropic turbulence and dimensional analysis, and derived the following relationship

$$E_L = K l^{4/3} e^{1/3}, \quad (1)$$

where K is a constant and l is an approximate diameter of the large vortices. The authors assumed that the vortices are responsible for the liquid mixing, and that their sizes being as large as the column diameter D . Since in bubble columns the energy dissipation e is equal to a product of g and u_G , therefore Eq. (1) can be written in the form

$$E_L = K D^{4/3} (g u_G)^{1/3}. \quad (2)$$

As seen, the dispersion coefficient depends on the column diameter as well as on the gas phase velocity. These findings were confirmed in the experimental studies of many other research groups.⁷⁻⁹ Eq. 2 can be rearranged

$$\frac{E_L}{D u_G} = K \left(\frac{g D}{u_G^2} \right)^{1/3} \quad (3)$$

and written in terms of the dimensionless Bodenstein and Froud numbers as follows

$$Bo_L^* = \frac{1}{K} (Fr^2)^{1/3}. \quad (4)$$

Eq. (4) with $K = 0.35$ encloses experimental data of many authors.

Whalley and Davidson¹⁰ imposed that a single circulation cell of the liquid is formed in a column. On the basis of this postulate Joshi and Sharma¹¹ correlated the liquid dispersion coefficient by means of the liquid phase circulation velocity

$$E_L = 0.031 D^{1.5} u_Z, \quad (5)$$

where the circulation velocity u_Z is given in cm s^{-1} , the column diameter D in cm , and E_L in $\text{cm}^2 \text{s}^{-1}$. The circulation liquid flow rate through the whole column length can be calculated from the relation¹²

$$u_Z = 14.4 [L(u_G - \varepsilon_G u_S)]^{1/3}. \quad (6)$$

For bubbles in a range of 2.4 - 8 mm, the bubble slip velocity u_S is about 23 cm s^{-1} , as reported by Joshi and Sharma.¹² For tall columns it is more realistic that circulation cells with the length of column diameter are formed. In this case the following expression for the u_Z is proposed¹²

$$u_Z = 13.9 [D(u_G - \varepsilon_G u_S)]^{1/3} \quad (7)$$

and the correlation for the dispersion coefficient becomes

$$E_L = 0.31 D^{1.5} u_Z. \quad (8)$$

Towell and Ackermann⁷ performed experiments with large diameter bubble columns and correlated their data in the form

$$E_L = 1.23 D^{1.5} u_G^{0.5}. \quad (9)$$

This expression confirms the Joshi and Sharma¹¹ theoretical approach, which stated that only two parameters affecting E_L values, they are the column diameter and the gas flow rate. Similar correlation was proposed by Badura et al.⁸ and later Deckwer et al.⁹ $E_L = 2.7 D^{1.4} u_G^{0.3}$. (10)

In developing the above correlation the authors incorporated data of many other investigators. The gas superficial velocity in Eq. (10) should be given in cm s^{-1} , D in cm , and coefficient E_L in $\text{cm}^2 \text{s}^{-1}$.

The correlations for predicting E_L in concurrent up-flow bubble columns filled with conventional packing are gathered in a review of Hoffman¹³, where the influence of the liquid flow rate as well as the packing dimensions on E_L are emphasized. From the correlation of Stiegel and Shah¹⁴

$$\text{Bo}_{L,p} = 0.128 \text{Re}_{L,p}^{0.245} \text{Re}_{G,p}^{-0.16} (a_s d_p)^{0.53} \quad (11)$$

one can conclude that E_L increases with the liquid flow rate and it is nearly insensitive to the gas flow rate. Smaller particles also diminish the liquid back-mixing in the column.

Experimental Apparatus

The experimental system used in this study is depicted in Figure 1. The column of circular cross-section was made of Plexiglas and had a 0.14 m ID and 1.885 m of total packing length.

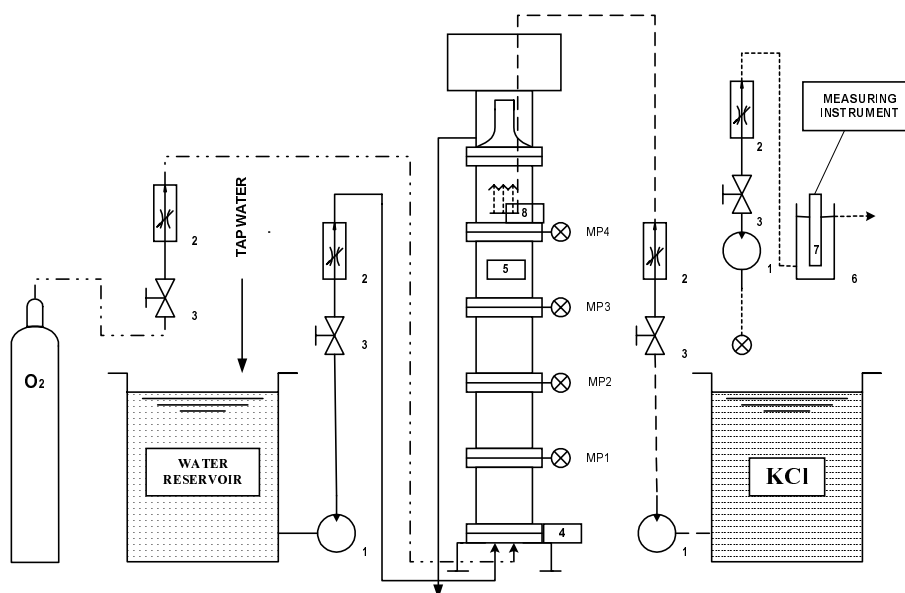


Figure 1. Experimental apparatus: 1.pumps, 2.rotameters, 3.regulating valves, 4.distributor, 5.column, 6.measuring cell, 7.electrode, 8.tracer injection device, ⊗ stopcocks.

The column consisted of five segments and was equipped with taps for collecting samples for measuring tracer concentration at different axial positions. The tracer concentrations were determined with a help of conductometric electrode. Tap water and oxygen were used as the contacting phases in concurrent up-flow flow mode. The water from the reservoir was introduced into the column through a hole of 2.5 cm ID at the column bottom. Due to relatively high h/D ratio a single-orifice (4.5 mm dia.) gas distributor placed 7 cm above the bottom was used for introducing oxygen into the column. Tests by a colored ink-tracer and a fast photo-technique showed that the single-orifice gas distributor did not affect the liquid flow neither in packed nor in non-packed column. The distance between the first packing element and the gas distributor was 2.0 cm. The centrifugal four-staged pump (KovinoTehna, Maribor) provided the liquid flow rates up to 3 m³/h. Flow rates of both phases were measured by means of calibrated rotameters. The column operated at atmospheric pressure while temperature of water in the column was kept at 12⁰C. The operating conditions are given in Table 1.

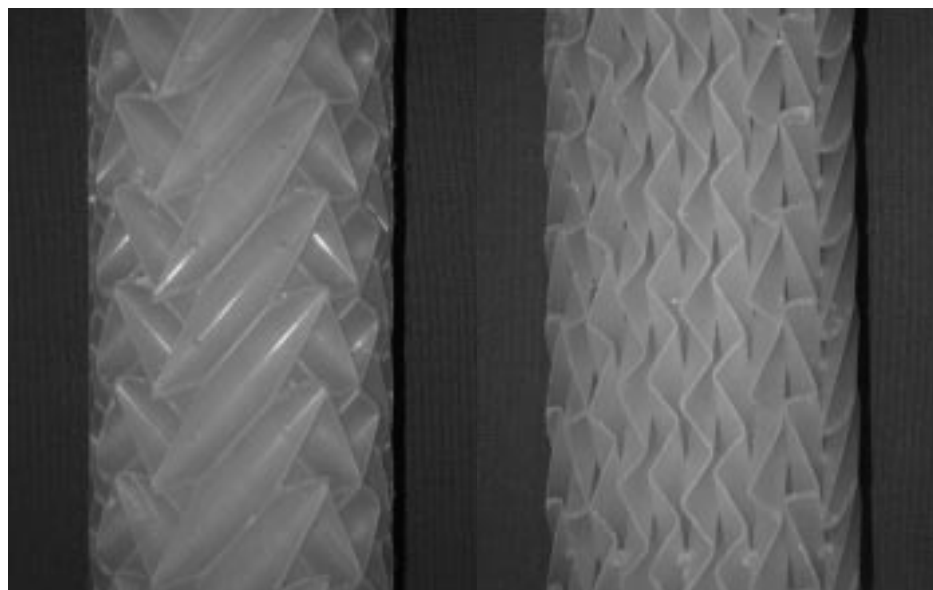
Table 1. Properties of the column and operating conditions

Column diameter	14.0 cm
Operating height	188.5 cm
Measuring points:	
MP 1	36.7 cm above the gas inlet
MP 2	74.4 cm above the gas inlet
MP 3	112.1 cm above the gas inlet
MP 4	149.8 cm above the gas inlet
Superficial velocity:	
Liquid (tap water)	0.36 - 5.41 cm/s
Gas (oxygen)	0.98 - 9.0 cm /s
Temperature of the liquid phase	12 ⁰ C ± 1 ⁰ C

The experiments were conducted in the column packed with five Sulzer SMV 16 elements (Figure 3) as well as in the column without packing. Between the packing elements spacing rings of 6.0 cm in length were inserted. The adjacent packing elements were rotated for an angle of 90⁰. The characteristics of the packing are listed in Table 2.

Table 2. Properties of packing

Material	Polyethylene
Diameter (mm)	140
Height (mm)	317
Void fraction (/)	0.85

**Figure 3.** The Sulzer SMV 16 packing elements (rotated for 90⁰)

Liquid phase dispersion measuring technique

Axial dispersion coefficient in the liquid phase is one-dimensional description of all mixing phenomena that occur on the macroscopic scale and is usually sufficient to describe the non-ideal flow of the liquid phase in bubble column. The steady-state method was employed in this experimental study. Although this is a very time consuming method, it is much more reliable than the non-steady-state (pulse) technique. Experimental arrangement of this method is illustrated in Figure 4, while a description of the method one can find elsewhere.¹ In the case of concurrent up-flow of both phases, the tracer substance was continuously fed into the liquid phase above the sampling taps. The back-mixing phenomena distribute the tracer along the column in the opposite direction of the liquid flow. Continuous sampling of the liquid along the column axis enables the determination of tracer concentration profiles under the steady-state

conditions. In these experiments a 0.2 M solution of KCl was used as tracer substance. The tracer was fed into the column by means of a peristaltic pump (Cole-Parmer Masterflex) at the axial position of 1.55 m from the gas distributor (position x_s in Figure 4) through an injection device. The device was simply a perforated ring with external diameter of 7.7 cm and with 24 holes of a 1 mm dia. Volumetric flow rates of the tracer solution was kept at 2% of the total liquid flow through the column.

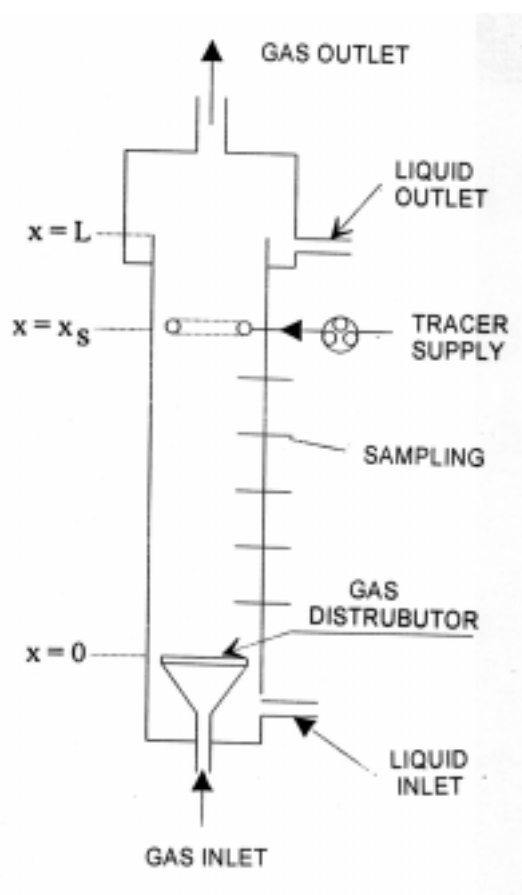


Figure 4. Schematics of the setup for experimental determination of liquid axial dispersion coefficient by the steady-state method.

Evaluation of liquid phase axial dispersion coefficient

The tracer injection device should evenly distribute the tracer across the column, therefore one-dimensional flow of the liquid phase can be assumed. Thus, the mass balance of tracer over the column differential element for the concurrent arrangement takes the form (see Figure 5)

$$E_L \frac{d^2 c_L}{dx^2} - u_L \frac{dc_L}{dx} = 0, \quad (12)$$

where the axial dispersion coefficient E_L is defined according to the analogy with the Fick's diffusion law, thus

$$J_D = -E_L \frac{dc_L}{dx}. \quad (13)$$

Deckwer¹ recommended the following boundary conditions for Eq. (12)

$$c_L = \frac{E_L}{u_L} \frac{dc_L}{dx} \quad \text{for} \quad x = 0, \quad (14)$$

$$c_L = c_0 \quad \text{for} \quad x_S \leq x \leq L. \quad (15)$$

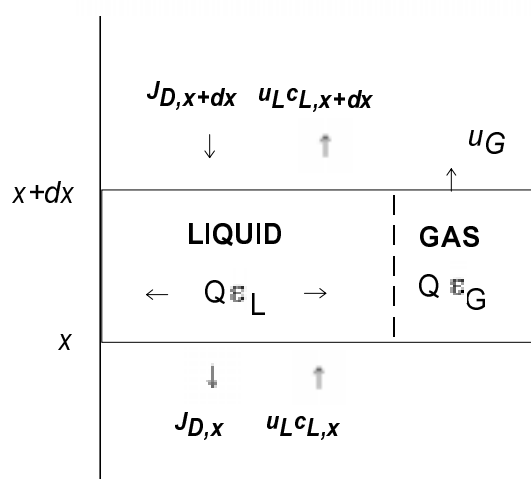


Figure 5. Convective and dispersive fluxes in column differential element.

The solution of Eq.12 with the above boundary conditions (Eq. 14 and Eq. 15) yields the following concentration profile

$$c_L = c_0 \exp \left[-\frac{E_L}{u_L} (x_S - x) \right]. \quad (16)$$

Once the tracer concentration profile at given operating conditions is experimentally determined, the value of E_L is calculated from the slope in plot concentration (logarithmic scale) vs. the column length.

Gas and liquid holdups

Gas and liquid holdups were measured by recording the levels (h_0) of aerated and non-aerated (h) liquid in the column. The gas and liquid holdups were then calculated according to the relations

$$\varepsilon_G = \frac{h_0 - h}{h_0}, \quad (17)$$

$$\varepsilon_L = \frac{h}{h_0}. \quad (18)$$

Results and discussion

The resulting liquid axial dispersion coefficients as a function of the gas superficial velocity in non-packed column are shown in Figure 6. The values are found between 86 and 257 cm^2s^{-1} but one can hardly conclude that there is some pronounced effect of the liquid flow rate on E_L . Scattering of the data can be mainly ascribed to the non-uniform flow with respect to the radial coordinate of the column. Except for one set of the data ($u_L=0.36 \text{ cm s}^{-1}$), the dispersion coefficients increases with the gas superficial velocity, which is in agreement with other investigators.^{2,3,6-8} At the lowest liquid velocity the measured E_L exhibits a maximum value at u_G of about 4 cm s^{-1} . Rice et al.¹⁵ also observed this phenomenon at low liquid flow rates. They explained this as a result of the hydrodynamic regime, which can be very sensitive not only to u_G , but also to u_L and the type of gas distributor. According to Deckwer¹ this experimental study did cover two different flow regimes: at low gas velocities (up to 3 cm s^{-1}) the bubbles formed were nearly uniform in size and they rose at almost constant velocity. This is called the homogeneous or bubble flow regime. When the gas velocities were increased, heterogeneous regime appeared. Due to the bubble coalescence large bubbles were formed and they traveled faster through the liquid. In general, this occurs at u_G higher than 5 cm s^{-1} . The transition between the two regimes somewhat depends on the type of gas distributor and material properties.

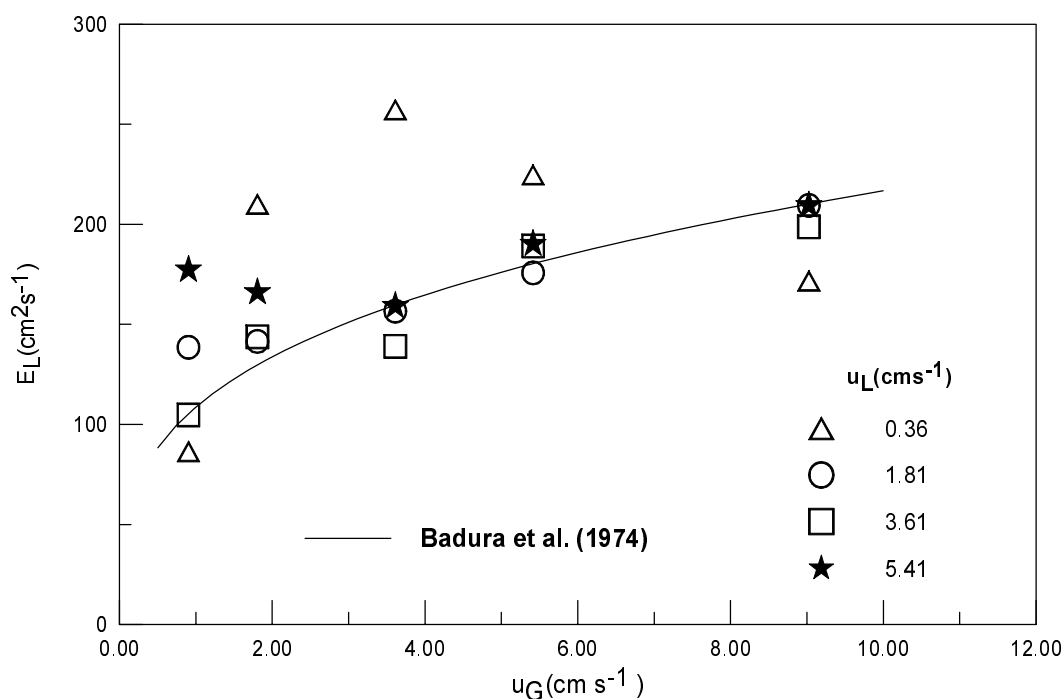


Figure 6. Liquid axial dispersion coefficient as a function of the gas superficial velocity in bubble column without packing.

In order to evaluate our results for structural packing, the experimental results were confronted with recommended correlations for E_L in non-packed columns. Thus, Joshi and Sharma models,^{11,12} which are based on the circulation cells, gave nearly 50% lower values compared to our data. Towell and Ackerman correlation⁵ also underestimates the axial dispersion coefficients. It seems that the best agreement with our experimental data is obtained with the correlation proposed by Badura et al.⁷ (see Table 3).

Table 3. Liquid axial dispersion coefficient prediction (statistics)

Reference	e_z (%)	σ (%)
Joshi and Sharma (1978)	45	51
Joshi and Sharma (1979)	40	65
Towell and Ackerman(1972)	30.3	18.9
Badura et. al (1974) ⁶	14.1	13.5

The effect of liquid flow rate is further diminished if the modified Bodenstein number (modified by gas superficial velocity and column diameter as a linear dimension) is plotted against the gas superficial velocity (Figure 7). As can be seen in Figure 7, the theoretical relation of Baird and Rice,⁶ which is based on the Kolmogoroff

theory, agrees very well with our experimental data (within $\pm 19\%$; $\sigma = 16.5\%$).

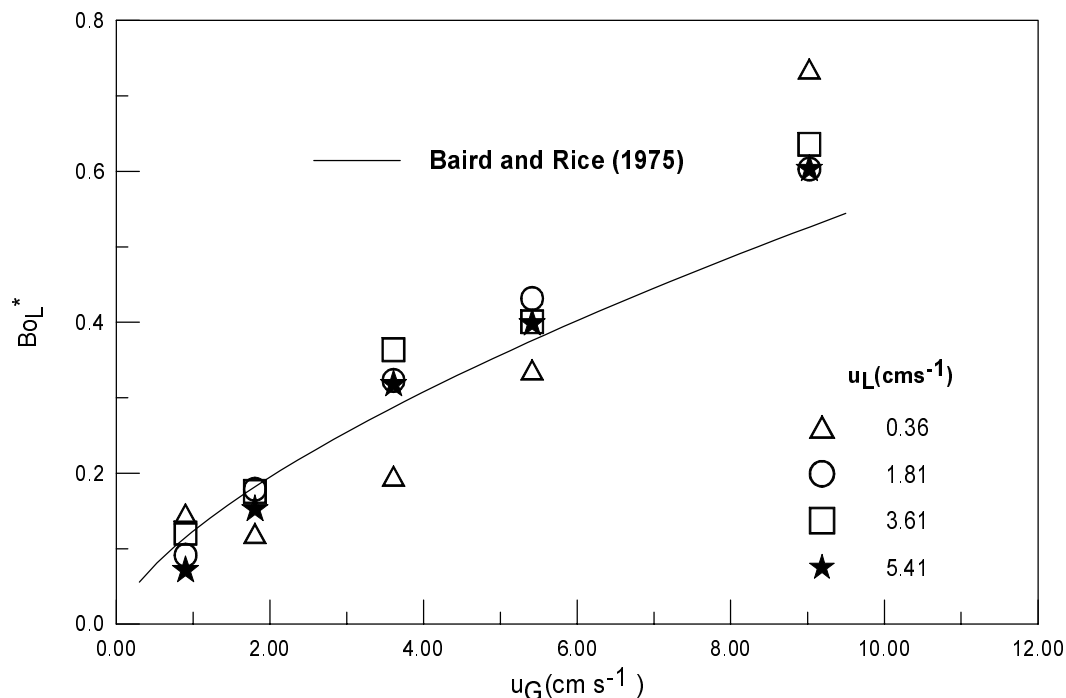


Figure 7. Modified Bodenstein liquid number as a function of the gas superficial velocity in bubble column without packing.

Figure 8 shows the results obtained in the packed column. Only a weak dependence on the gas superficial velocity was observed, but it doubled when the liquid velocity was raised from 0.38 cm s^{-1} to 5.41 cm s^{-1} . The experimental error in the E_L determination was much higher (up to 30 %) compared to the results of non-packed column (10 %). Maclean et al.¹⁶ who studied the oxygen transfer in bubble column containing static mixers also reported the experimental error of the same range. Contrary to the results found in the non-packed column no maximum with the axial dispersion coefficient as a function of the gas velocity appeared in the column with structural packing. Spicka et al.¹⁷ experimentally verified that the presence of packing extends the range of the bubble flow regime even up to 9 cm/s of the gas superficial velocity. Therefore one can reasonably conclude that the experiments performed with structural packing in this work lied in the homogeneous regime. The liquid axial dispersion coefficient was found to be proportional to the liquid superficial velocity to a power of 0.3.

However, a comparison of the liquid axial dispersion coefficients obtained in two different bubble column arrangements is shown in Figure 9 as a function of liquid velocity. Even though the data points are somehow scattered, one can conclude that the

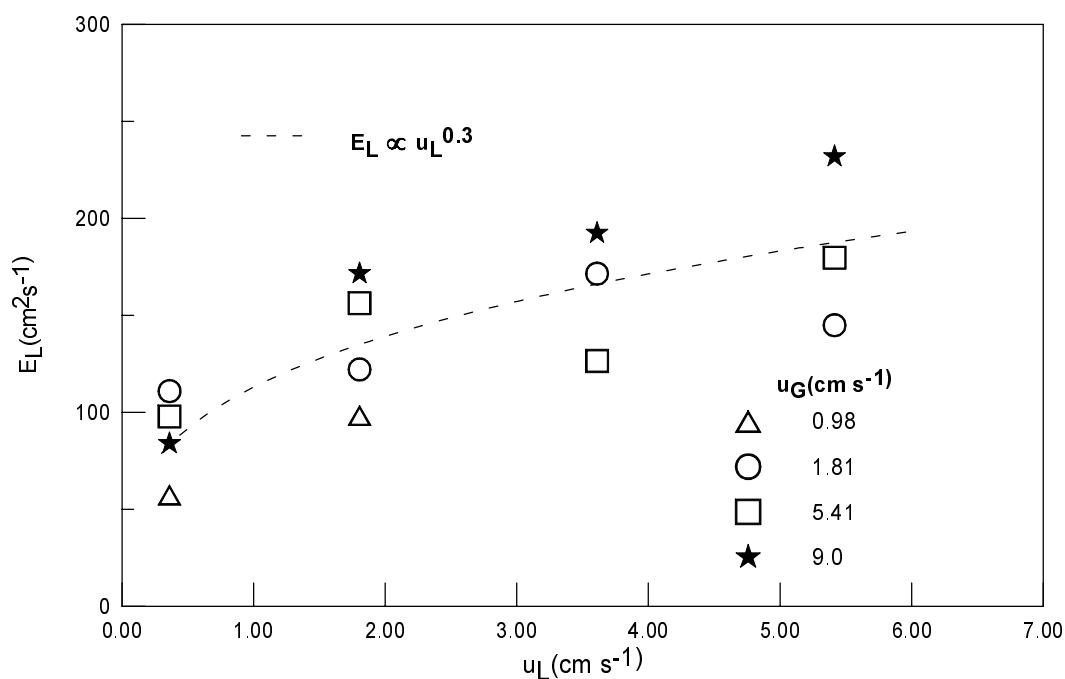


Figure 8. Liquid axial dispersion coefficient as a function of the liquid superficial velocity in bubble column with structured packing.

liquid axial dispersion coefficient increases with the liquid flow rate in the column packed with the structured packing, whereas in the non-packed column it is practically constant. Thus, the presence of Sulzer SMV 16 packing lowers the axial dispersion coefficient in the liquid: it is for about 50 % at low gas flow rates but only for 20 % at high gas throughputs. An average value of the liquid axial dispersion coefficient in the non-packed column is estimated to be about $172 \text{ cm}^2 \text{ s}^{-1}$.

The dependence of the Bo_L on the liquid velocity is shown in Figure 10. Here the Bodenstein number for the liquid phase is based on the liquid superficial velocity and the column diameter. As it can be seen from the figure, the experimental data plotted with gas flow rate as a parameter split at higher liquid velocities. Higher gas flow rates yield lower values of Bo_L , which is in agreement with the results of Maclean et al.¹⁶

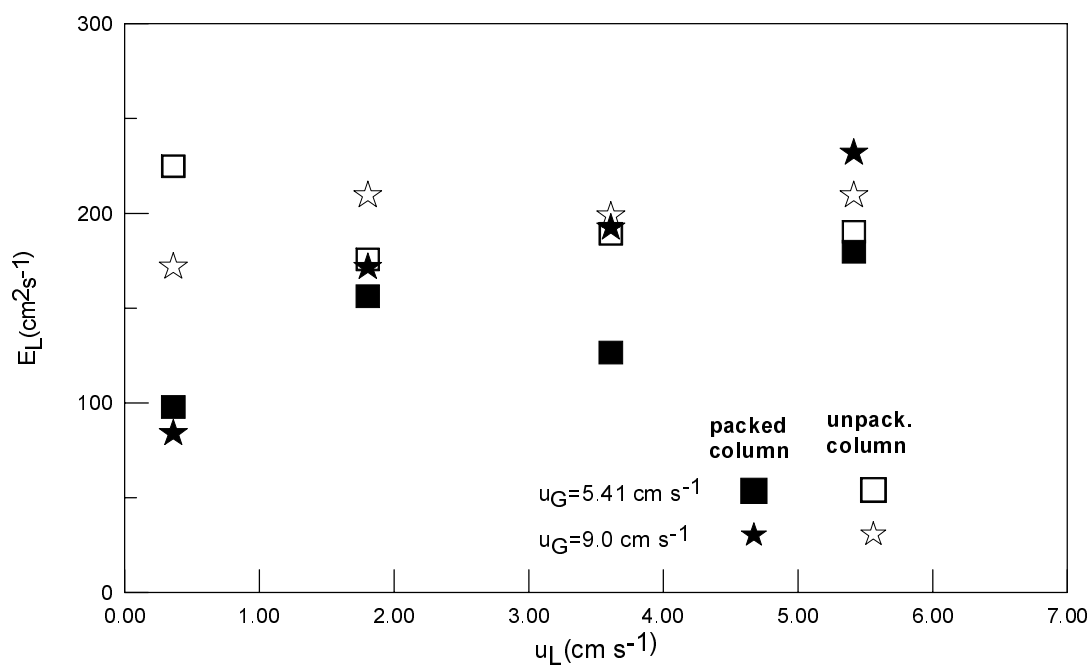


Figure 9. Comparison of liquid axial dispersion coefficients obtained in two different arrangements of bubble column.

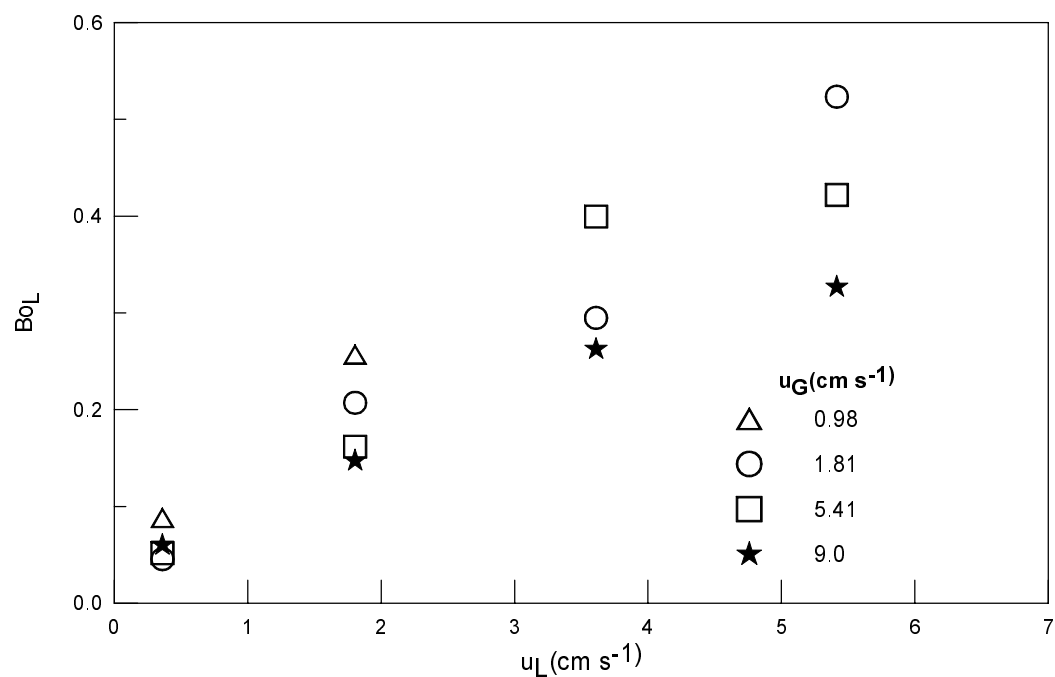


Figure 10: Modified Bodenstein liquid number as a function of the liquid superficial velocity in bubble column with structured packing.

Conclusion

The axial dispersion coefficients in bubble column packed with Sulzer SMV 16 are lower for about 50 % at low gas flow rates and only for 20 % at high gas rates. The gas flow rate increases the E_L values in both cases, in packed and non-packed bubble columns, whereas the liquid superficial velocity affects the axial mixing only in column with structural packing.

Acknowledgement

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Nomenclature

a_s	specific surface area of internals, m^2m^{-3}
Bo_L	Bodenstein liquid number, $\left[= \frac{u_L D}{E_L} \right]$, /
Bo_L^*	modified Bodenstein liquid number, $\left[= \frac{u_G D}{E_L} \right]$, /
$Bo_{L,p}$	Bodenstein liquid number, $\left[= \frac{u_L d_p}{E_L} \right]$, /
c	concentration of the tracer, gm^{-3}
d_p	particle diameter, m
D	column diameter, m
e	energy dissipation, m^2s^{-3}
e_y	mean relative error, $\left(= \frac{100}{N} \sum_{i=1}^N \left \frac{y_{MES}(i) - y_{PRED}(i)}{y_{MES}(i)} \right \right)$, %
Fr	Froude number, $\left[= \frac{u_G}{g D} \right]$, /
E_L	dispersion coefficient, m^2s^{-1}
g	acceleration due to gravity, ms^{-2}
h	operating height of the aerated column, m
h_0	gas-free liquid level, m
J_D	dispersion flux, $gm^{-2}s^{-1}$
N	number of experimental data
Re_p	Reynolds particle number, $\left[= \frac{u d_p \rho}{\eta} \right]$, /
u	superficial velocity, ms^{-1}

Q column cross section area, m²

Greek letters

ε phase hold-up, /

μ viscosity of the phase, kgm⁻¹s⁻¹

ρ density of the phase, kgm⁻³

σ standard deviation, $\left(= 100 \sqrt{\frac{1}{N-1} \sum_{i=1}^N \left[\frac{y_{\text{PRED}}(i) - y_{\text{MES}}(i)}{y_{\text{MES}}(i)} - e_y \right]^2} \right)$, %

Subscripts

G gas phase

MES measured value

L liquid phase

o entrance of the column

p particle

PRED predicted value

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Povzetek

Podani so rezultati študije vpliva strukturiranega polnila na stopnjo aksialnega mešanja kapljevinate faze v kolonah-reaktorjih z mehurčki. Kot polnilo so bili uporabljeni statični mešalniki firme Sulzer (SMV 16). Za primerjavo so bili izvedeni tudi eksperimenti v koloni brez polnila. Izkazalo se je, da vrednosti koeficienta aksialne disperzije naraščajo s hitrostjo plinaste faze v obeh tipih kolon, vendar pa pretok kapljevine rahlo zvišuje vrednosti koeficienta le v koloni s polnilom. Vrednosti koeficienta v primeru uporabe strukturiranega polnila so za okoli 50 % nižje pri visokih hitrostih plina in le za okoli 20 % pri nizkih hitrostih v primerjavi z onimi v koloni brez polnila.