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Mass transfer to the wall of a packed and unpacked bubble column operating with Newtonian and non-Newtonian liquids

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Abstract

Mass transfer to the wall of a bubble column is examined, focusing on the influence of the liquid viscosity. The effect of introducing structured packing in the column is analysed by comparing results for the empty column with those of packed column at similar operating conditions. Visual observation allowed detecting of flow transition in the empty column with non-Newtonian fluids; hence, data obtained in a different flow regime are correlated separately. Results from the empty and packed columns could be correlated by the same expression, indicating that the presence of the structured packing inside the bubble column has no remarkable influence on the liquid to wall mass transfer. Expressions of the form $St = f(Re, Fr, Sc)$ or $Sh = f(Sc, Ga, E)$, mostly used in investigations on heat or mass transfer in bubble columns, are proposed.

Keywords: Bubble columns; Structured packing; Mass transfer; High viscosity

1. Introduction

Bubble columns are supplemented with structured packing in the process industry due to many reasons. In the ever growing field of biotechnology, micro-organisms can be fixed on the packing surface [1], or structured packing may be used as catalyst support in chemical reactors [2,3]. Their presence reduces the undesirable back-mixing of the involved phases, arising from the internal fluid circulation that characterises the empty bubble columns [4]. Structured packings are also employed as static mixers, replacing successfully other stirring devices. For instance, in bioreactors the low shear imposed prevents damaging sensitive cells during processing, as frequently happens with moving agitators. Also, specially with high viscosity and non-Newtonian liquids, static mixers produce a uniform velocity profile over the whole cross section, while the velocity decays from the centre to the column wall with rotating agitators. The gas holdup distribution is also even due to their excellent radial mixing performance [5]. Furthermore, the presence of regular packings enhances gas holdup compared to empty bubble columns [5–7]; although the opposite behaviour was found for highly viscous liquids [8].

When regular packings are inserted in a bubble column carrying out reactions under controlled conditions, heat may be exchanged through the column walls [9]. When the bubble column reactor is made up of several structured sections, the heat evolved due to the reaction can also be removed by external heat exchange [4].

The use of static mixers to enhance heat transfer through the pipe wall of heat exchangers with flowing fluids is well known and has been extensively investigated, both by experiments [10] and by computational fluid dynamics [11]. Heat transfer coefficients were found to be several times higher than those corresponding to empty pipes for laminar and turbulent flow and also in the transition region.

Heat transfer improvement in a catalytic reactor by insertion of a special type of structured packing in the reactor tube has been investigated for catalysed gas phase reactions [12]. Heat transfer coefficients for the inner tube wall increased about 20% in comparison to beds of random particles or commercially available static mixers. However, to the authors' knowledge there is no information in the literature on the behaviour of heat exchanging in bubble columns containing static mixers (or regular packing). Due to the analogy that holds for heat and mass transport phenomena, the fluid to wall mass transfer measurements could yield useful insights for the design of bubble column reactors requiring heating or cooling. In this work, mass transfer towards the bubble column wall is examined, both for an empty bubble

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Nomenclature

a	surface area per unit volume (m^{-1})
d	bubble diameter (m)
D	diffusivity ($m^2\ s^{-1}$)
E	gas void fraction
Fr	Froude number = $Vs^2\ g^{-1}\ d^{-1}$
g	gravitational acceleration ($m\ s^{-2}$)
Ga	Galileo number = $g\ L^3\ \rho^2\ \mu^{-2}$
H	clear liquid height (m)
H_D	gas–liquid dispersion height (m)
h	height of packing element (m)
k	mass transfer coefficient ($m\ s^{-1}$)
K	consistency index ($Pa\ s^n$)
L	electrode length (m)
n	flow index
Re	Reynolds number = $Vs\ d\ \rho\ \mu^{-1}$
Sc	Schmidt number = $\mu\ \rho^{-1}\ D^{-1}$
Sh	Sherwood number = $k\ L\ D^{-1}$
St	Stanton number = $k\ Vs^{-1}$
Vs	superficial gas velocity ($m\ s^{-1}$)

Greek letters

α	folding angle of furrows (°)
β	corrugation inclination angle (°)
ε	packing void fraction
μ	dynamic viscosity (Pa s)
ρ	liquid density ($kg\ m^{-3}$)

Subscripts

ap	apparent
ec	empty column
p	packed column

column and for the column packed with structured packings. Furthermore, since viscous media are often used in this type of gas–liquid contactor, special attention is paid to the bubble column behaviour with viscous and non-Newtonian liquids.

2. Experimental set-up and methods

The experiments were carried out in a square section acrylic column (4 cm × 4 cm, 80 cm high) equipped with two to five cubic regular packing elements of the type Sulzer-Mellapak. Fig. 1 shows a schematic diagram of the experimental set-up.

As usual, consecutive packs were stacked in the column rotated 90° in between. Essential parameters of the ten corrugated sheets composing each regular packing element are: the height of pack $h = 4$ cm, the channel inclination angle $\beta = 30°$, the folding angle of the furrows $\alpha = 97°$, the channel side $s = 0.567$ cm, the surface area per unit volume $a = 8.23\ cm^{-1}$ and the void fraction $\varepsilon = 0.88$. The two lower packs were made of pure nickel; the other elements were made in plastic. Fig. 2 shows a photo of the metallic and plastic packing elements.

A perforated plate with 16 holes of 0.5 mm, in combination with the two metallic elements of regular packing, worked as gas sparger [13]. Humidified nitrogen was used, with the superficial velocity ranging from 0.006 to $0.08\ m\ s^{-1}$. Experiments were carried out in steady state conditions. The manometric and displacement techniques were employed to determine the fractional gas holdup in the column. In the first case, the hydrostatic pressure was measured using pressure taps at 2 and 18 cm above the sparger. With the displacement technique, the gas holdup was estimated from the liquid height without flowing gas, H (equal to 25 cm in all experiments) and the height of dispersion H_D:

$$E = \frac{H_D - H}{H_D} \quad (1)$$

For the high viscosity fluids, only the displacement method could be applied since the motion of high viscous liquids through the narrow manometer tubes is difficult. It is worth mentioning that with these liquids, an accumulation of very small stagnant bubbles was observed.

All experiments were carried out at room temperature and batch-wise with respect to the liquid. Since mass transfer coefficients were obtained by the electrochemical method, all the solutions contained the redox pair potassium ferri/ferrocyanide and a supporting electrolyte.

Carboxymethyl cellulose sodium salt was added to the aqueous solutions (except for solutions S1 and S2), in order to modify their viscosity. For high CMC-concentrations, non-Newtonian behaviour was found, characterised by the Ostwald- de Waele relation. The diffusion coefficients of the ferricyanide ion were obtained with a pipe wall electrode, as described by Tonini et al. [14].

At the beginning and at the end of each series of experiments, the rheological parameters and densities of the solutions were measured with a cone/plate viscometer and a pycnometer, respectively. No significant variation was detected. The composition and physical properties at 25 °C of the different electrolytic solutions employed are given in Table 1.

For measuring mass transfer coefficients from the fluid to the column wall by the limiting current method, three pairs of nickel electrodes (1.5 × 1.5 cm) flushed to opposite sides of the column wall were used. Electrodes were located at 8.6, 10.4 and 14.2 cm from the bottom and acted as test electrodes. The nickel packs resting on the perforated plate worked as the counter electrode. With these six cathodes, individual limiting current intensities were recorded and, since no important effect of the electrode position was observed, the mean value of the current intensity was used in the calculation of mass transfer coefficients.

Experiments with and without the plastic packing elements were performed for each electrolyte at different superficial gas velocities, yielding fluid to wall mass transfer rates for the packed and for the empty bubble column.

3. Results and discussion

3.1. Gas holdup

The gas holdup is an important design parameter in bubble columns and it is often used to describe the fluid dynamic characteristics in liquid to wall mass transfer correlations [15].

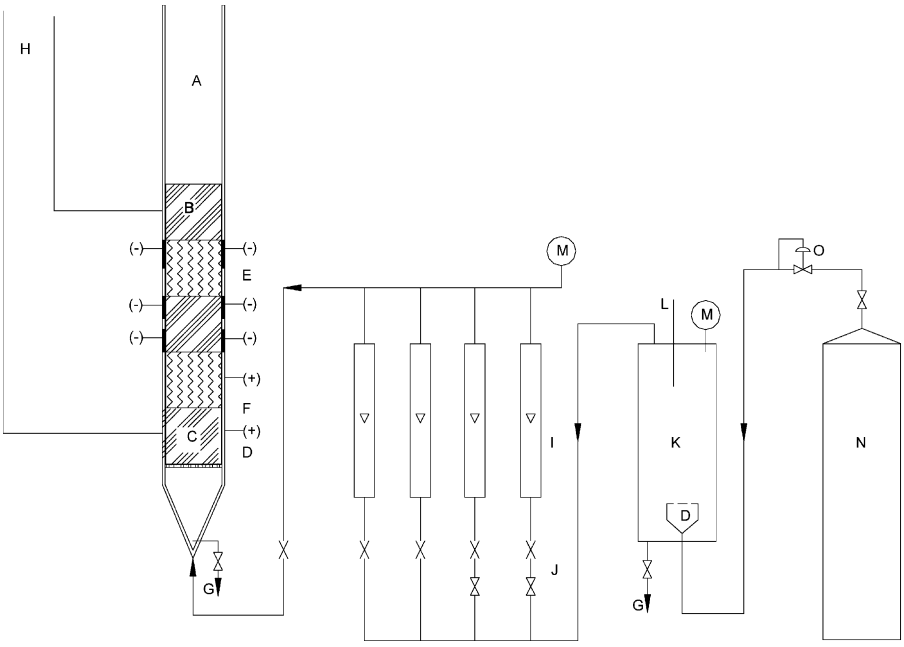

Fig. 1. Experimental set-up employed. (A) Bubble column, (B) plastic structured packing, (C) metallic structured packing, (D) gas spargers, (E) cathodes, (F) counter electrode, (G) drain, (H) open tube manometer, (I) rotameters, (J) valves, (K) gas saturator, (L) thermometer, (M) manometers, (N) nitrogen tube, (O) pressure regulator.

Measured overall gas void fractions are shown in Fig. 3a and b as a function of the superficial gas velocity, for the packed and the empty column, respectively.

The behaviour of both devices is clearly different. In the packed column, the gas holdup increases continuously with liquid viscosity and the gas flow rate. While working with non-Newtonian liquids, numerous micro-bubbles appear, leading to abnormal values of E at low superficial gas velocities. At high gas flow rates, the influence of the liquid viscosity clearly diminishes and E tends to a constant value as the gas velocity increases. In the empty column, for gas velocities below 2 cm s^{-1} and with Newtonian fluids, the gas holdup is barely sensitive to viscosity variations. Moreover, gas holdup values are always smaller than those measured in the packed bubble column at similar operating conditions. With non-Newtonian liquids slugs appear, producing a sharp increase of gas holdup. A gradual variation with fluid viscosity and superficial gas velocity is observed. Also, for the solutions with the same rheological parameters, higher values of gas holdup are found in the empty column compared to the packed column.

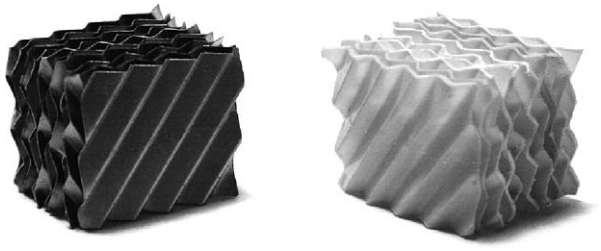

Fig. 2. Structured packing elements.

The same behaviour was observed in a previous investigation on gas–liquid dispersions in cylindrical bubble columns containing structured packings [8]. Besides the effect of the fluid

Table 1
Composition and physical properties at 25 °C of employed solutions

Solution	$[Fe(CN)_6^{3-}]/[Fe(CN)_6^{4-}]$ (kmol m^{-3})	[CMC] (Fluka) (%)	$\rho \times 10^{-3}$ (kg m^{-3})	μ or K (mPa s^n)	n (−)	$D \times 10^{10}$ (m^2 s^{-1})
S1	0.025	–	1.030	0.95	1	7.84
S2	0.010	–	1.060	1.20	1	5.51
S3	0.010	0.3 (low)	1.065	2.80	1	5.99
S4	0.010	0.6 (low)	1.066	5.06	1	5.78
S5	0.010	0.8 (low)	1.069	7.42	1	6.18
S6	0.010	1.0 (low)	1.069	10.57	1	6.22
S7	0.010	1.5 (low)	1.069	22.10	0.995	6.35
S8	0.001	1.5 (BDH)	1.070	67.00	0.963	6.47
S9	0.010	1.0 (high)	1.072	324.00	0.842	6.78
S10	0.010	1.25 (high)	1.072	958.00	0.814	6.90
S11	0.010	1.5 (high)	1.072	1763.00	0.782	6.96

Supporting electrolyte: S1: NaOH 0.5 M; S2–S11: Na_2CO_3/$HNaCO_3$ 0.4 M.

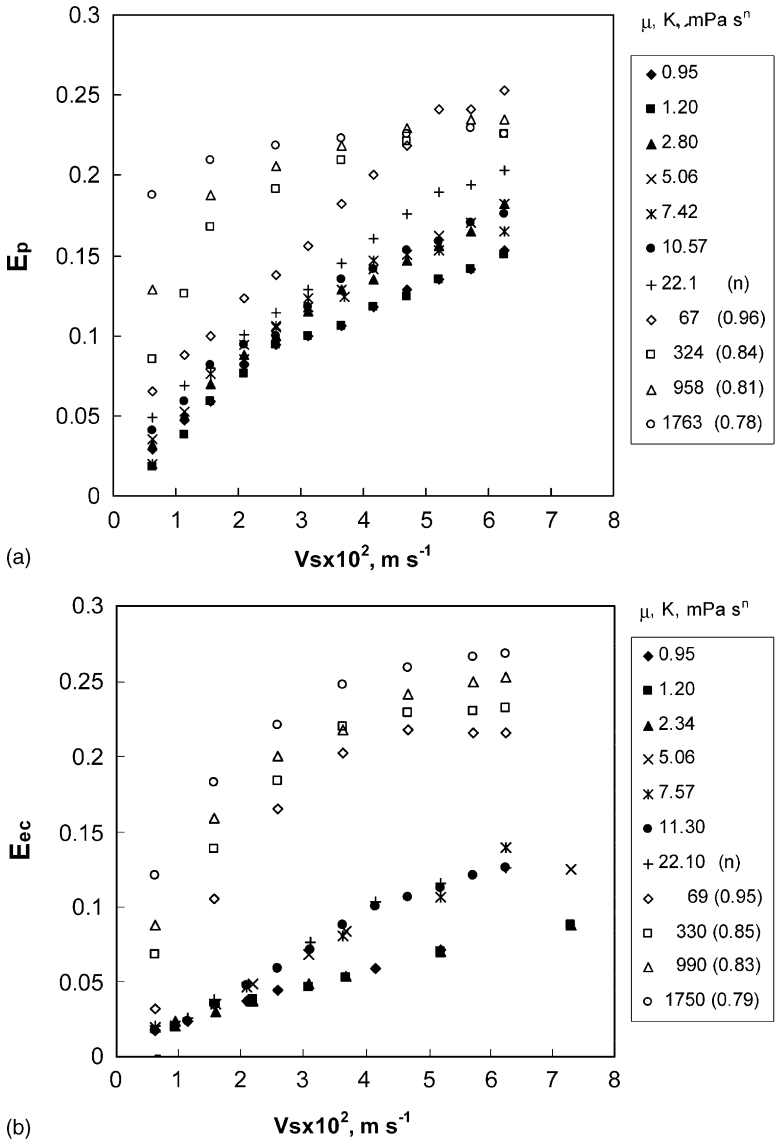

Fig. 3. Gas holdup in bubble columns for various liquid viscosities. (a) Column containing structured packing; (b) empty column.

viscosity on the gas holdup, the influence of the packing material had been studied. Equations describing the effect of liquid viscosity and gas superficial velocity on the gas holdup were proposed for the different packings and for the empty column, excluding in the latter system the data obtained under slugging flow regime. Thus, for the bubble column filled with a plastic structured packing similar to the one used in the present work and for the empty column, the following functions were developed:

$$E_{\mathrm{p}} = \frac{0.615\mu^{-0.098}\mathrm{Vs}}{\mathrm{Vs} + 20.54\mu^{-0.349}} \tag{2}$$

$$E_{\mathrm{ec}} = \frac{0.357\mu^{0.083}\mathrm{Vs}}{\mathrm{Vs} + 10.21\mu^{0.051}} \tag{3}$$

with Vs expressed in cm s^{-1} and μ in mPa s. In case of the aerated non-Newtonian fluids, the apparent viscosity calculated with the expression derived in [8] has to be employed:

$$\mu_{\mathrm{ap}} = K\left(\frac{Vsg\rho}{K}\right)^{(n-1)/(n+1)} \tag{4}$$

Fig. 4 compares the present experimental results of gas holdup to estimated values calculated with Eqs. (2) and (3). As can be seen from these parity plots, in presence of structured packing Eq. (2), derived with cylindrical columns, predicts the gas holdup in square columns quite accurately. Deviations higher than 25% between observed and predicted data only appear for low gas flow rates and high viscosity liquids. The same occurred in the circular packed column and is due to the micro-bubbles, not accounted for in the correlation. The fluid dynamic characteristics within the inclined channels of the packing are independent from the surrounding column geometry. For the empty column, within the same viscosity range as the one used in [8] developing Eq. (3), there is no coincidence between gas holdup values for circular and square columns. In the square column, secondary

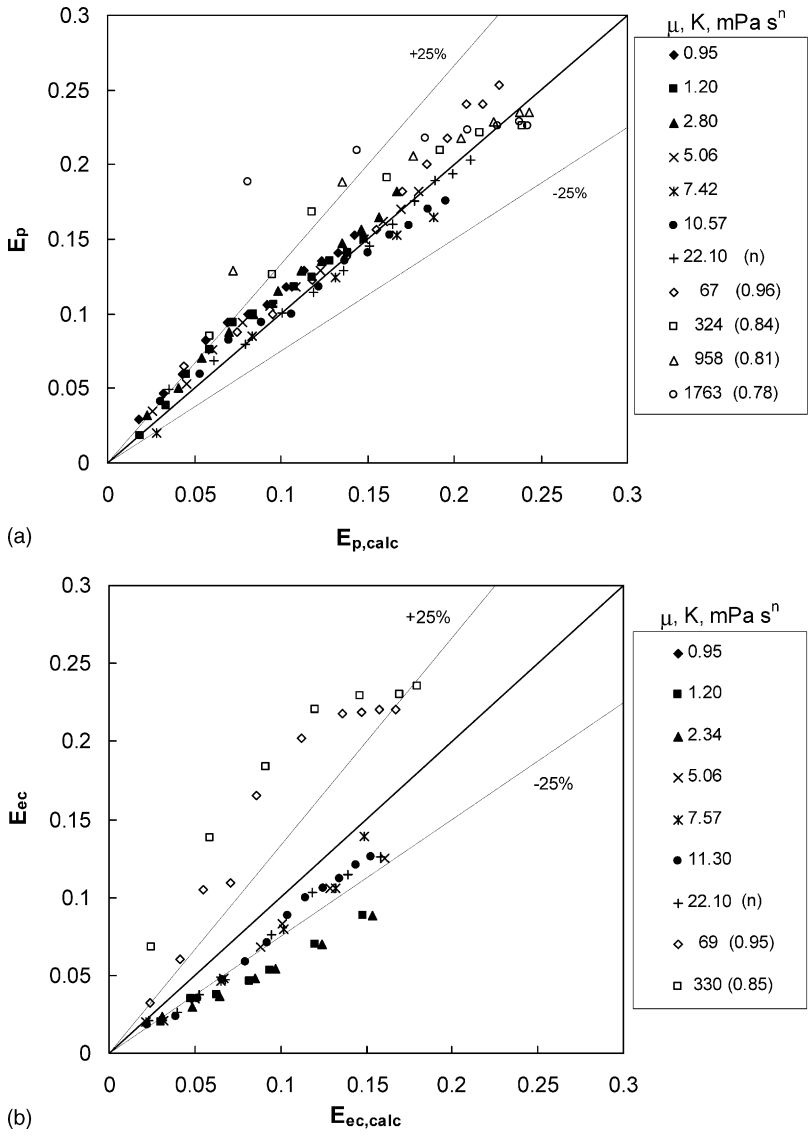

Fig. 4. Comparison of measured gas holdup with calculated values (a) packed column, Eq. (2); (b) empty column, Eq. (3).

flow effects appear in the corners, altering the flow pattern and hence the overall gas holdup.

3.2. Liquid to wall mass transfer

According to different models and correlations given in the literature for the prediction of heat or mass transfer in bubble columns, the parameter chosen to describe the fluid dynamics can be the gas holdup or the superficial gas velocity [15]. Therefore, mass transfer coefficients obtained for the packed and the unpacked column are presented in Fig. 5a–d as a function of superficial gas velocity and of gas holdup, respectively. Fig. 6 gives the relation between mass transfer coefficients obtained with and without structured packing for the same superficial gas velocity and similar fluid viscosity. From these graphs, it can be concluded that both columns present an analogous variation of mass transfer coefficients with superficial gas velocity.

When working with low viscosity solutions, for the same gas flow rate, *k*-values are very similar with and without structured packing. Against the expectations, the improvement arising from the presence of the structured packing is merely a few percents.

With high viscosity solutions, the opposite occurs. In the empty column, where slug flow arises, the free downwards circulation close to the transferring wall and the agitation caused by the ascending slugs enhance mass transfer coefficients, as compared to the packed column.

On the other hand, for equal gas void fractions, the mass transfer coefficients in the empty column are, in general, somewhat higher than in the packed column. However, it should be pointed out that to reach the same gas void fraction, in the empty column much higher gas flow rates have to be used. Furthermore, it can be seen that gas void fractions in the empty column are limited to low values. Only with the non-Newtonian solutions, higher gas holdups can be obtained (Fig. 5c and d).

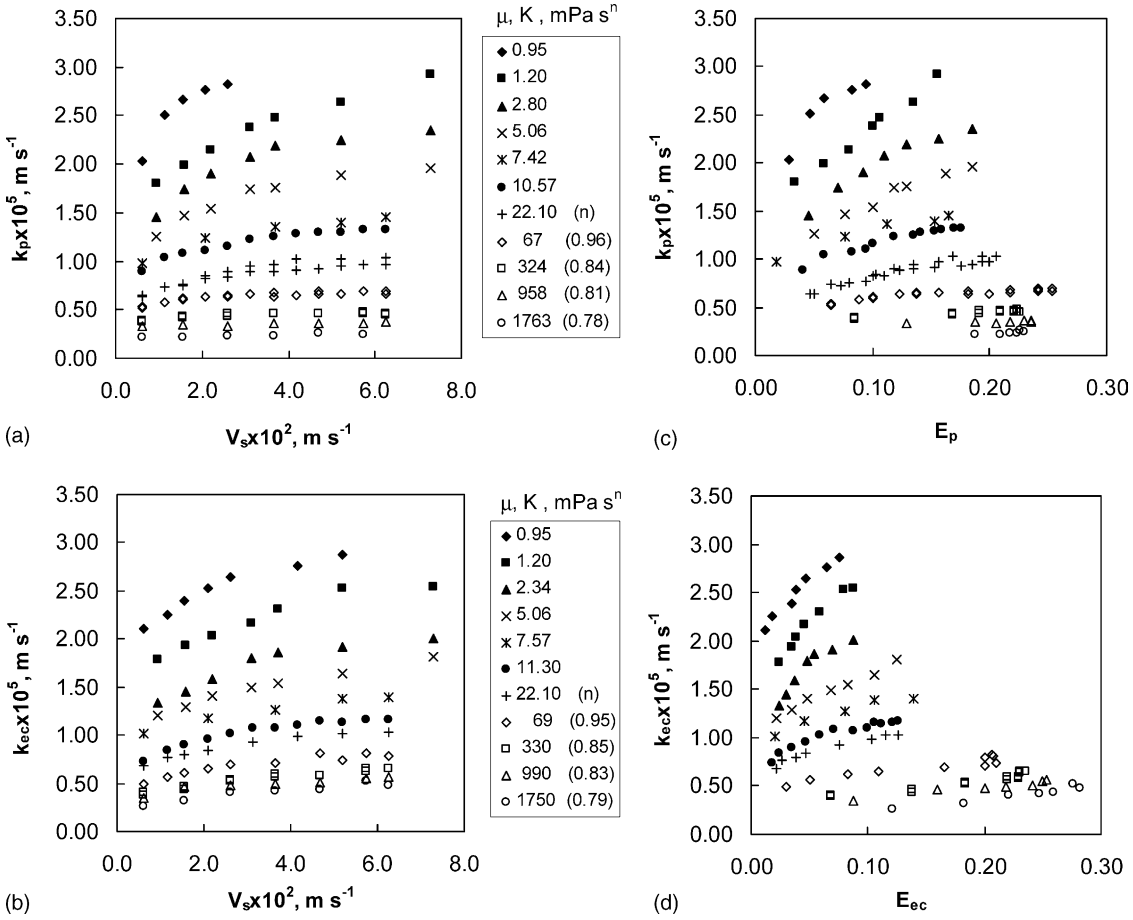

Fig. 5. Mass transfer coefficients vs. superficial gas velocity and versus gas holdup; (a) and (c) packed column; (b) and (d) empty column.

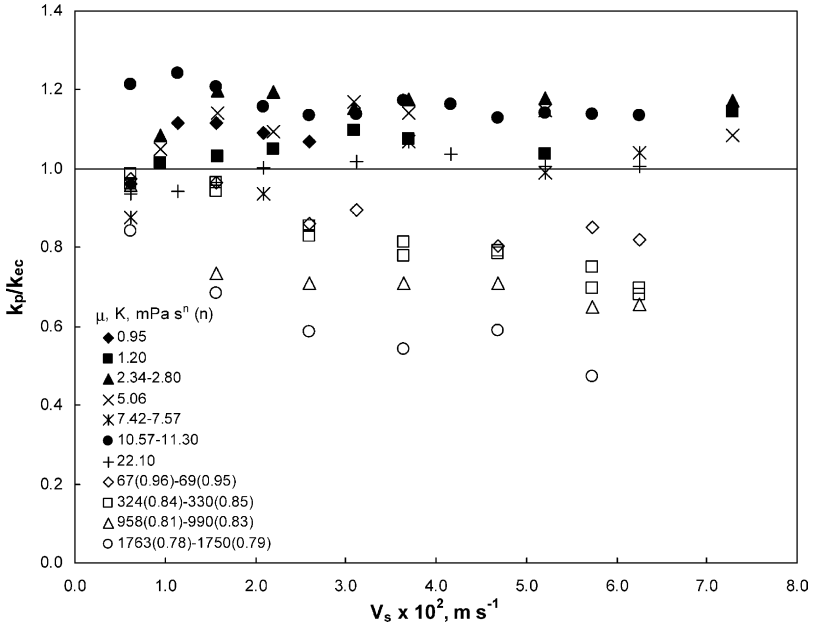

Fig. 6. Comparison of liquid to wall mass transfer coefficients in presence of structured packing and in the empty column.

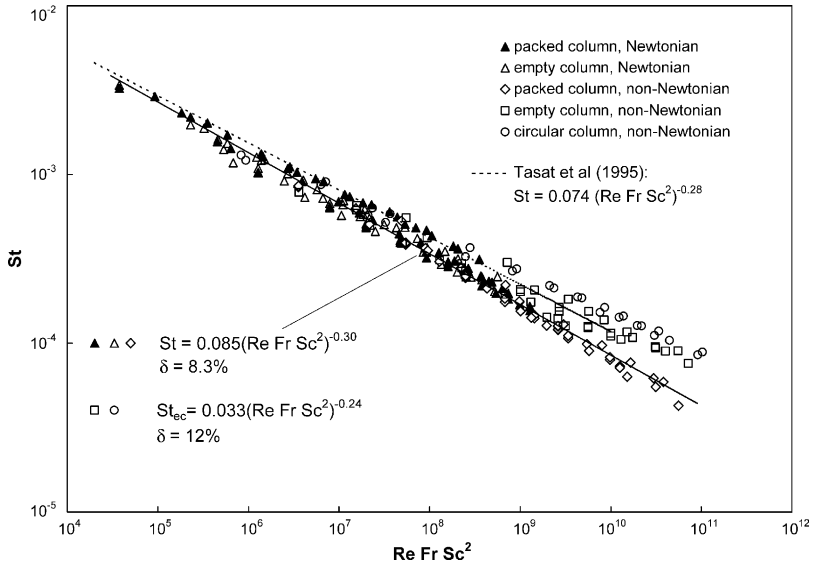

Fig. 7. General mass transfer correlation for empty bubble columns and columns containing structured packing.

4. Correlation of mass transfer coefficients

In order to correlate the liquid to wall mass transfer coefficients with the involved fluid dynamic variables and fluid physical properties, two approaches suggested in the literature were considered. One combines the surface renewal theory with Kolmogoroff's theory of isotropic turbulence [16], selecting the superficial gas velocity to describe the fluid dynamics. The other compares the motion of the gas–liquid dispersion with turbulent free convection [17] and makes use of the gas holdup to characterise fluid dynamics. Dimensionless relations of the type $St = f(Re, Fr, Sc)$ and $Sh = f(Sc, Ga, E)$ result, respectively. For the non-Newtonian liquids, the apparent viscosity given by Eq. (4) is introduced in the Reynolds, Schmidt and Galileo numbers.

In Fig. 7, all the mass transfer data are plotted as Stanton number versus ($Re\ Fr\ Sc^2$) (as it is commonly used in the literature). Some data for non-Newtonian solutions in an empty circular bubble column are also shown [18]. They were obtained by the same electrochemical technique used in the present work. The column was 5 cm i.d.; the test electrode was a nickel ring, 1.79 cm high, which replaced part of the column wall; the solutions were characterised by the power-law parameters $K = 483–536$ mPa s^n and $n = 0.87$.

Clearly the mass transfer results corresponding to non-Newtonian solutions in circular or square empty columns deviate from the general trend found with all the other results. Mass transfer coefficients are higher, likely due to the slug flow condition prevailing in these viscous media and are somewhat higher for the circular column than for the square one. The introduction of the packing hinders the formation of large slugs, limiting the size of the bubbles to that of the packing channels and no drastic flow transition occurs. Therefore, a separate correlation

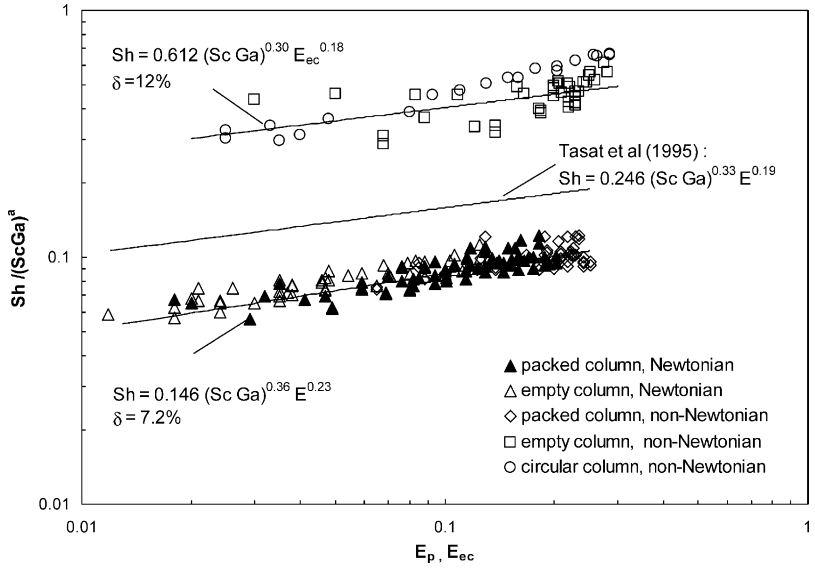

Fig. 8. General mass transfer correlation for empty bubble columns and columns containing structured packing.

of the data corresponding to non-Newtonian solutions in empty columns was developed:

$$St_{ec} = 0.033 \quad (Re \;\; Fr \;\; Sc^2)^{-0.24} \tag{5}$$

This empirical relation predicts the 64 available results for the slug flow regime with a mean error of 12%.

Since mass transfer coefficients do not vary appreciably with and without packing, all 170 data measured with Newtonian liquids in both systems and with power-law fluids in structured packing, were correlated with a single equation:

$$St = 0.085 \quad (Re \;\; Fr \;\; Sc^2)^{-0.30} \quad \delta = 8.3\% \tag{6}$$

Fig. 7 shows the correlating line corresponding to Eq. (6) together with a relationship proposed by Tasat et al. [7] to describe the liquid/solid mass transfer inside the structured packing:

$$St = 0.072 \quad (Re \;\; Fr \;\; Sc^2)^{-0.28} \tag{7}$$

This correlation was derived with Newtonian liquids and packing elements made of smooth nickel sheet. As can be seen, mass transfer coefficients for the corrugated plates in the interior of the packing are always higher than to the column wall, differences being about 40% at high values of ($Re\ Fr\ Sc^2$). This increase of mass transfer rate within the packing may be attributed to the existing interaction of crossing flows in the channels of adjacent packing plates.

Following the approach suggested by Sigrist et al. [17], the mass transfer coefficient is correlated to the gas void fraction and liquid physical properties through the Sherwood, Schmidt and Galileo numbers. As with the former model, the mass transfer results were subjected to statistical analysis separately. The following equations, represented in Fig. 8, give the best correlations of the experimental data:

For non-Newtonian liquids in empty columns:

$$Sh = 0.612 \quad (Sc \;\; Ga)^{0.30} E_{ec}{}^{0.18} \quad \delta = 13\% \tag{8}$$

For Newtonian and non-Newtonian liquids in the column with structured packing and Newtonian liquids in the unpacked column:

$$Sh = 0.146 \quad (Sc \;\; Ga)^{0.36} E^{0.23} \quad \delta = 7.2\% \tag{9}$$

Fig. 8 also includes the general correlation derived by Tasat et al. [7] for mass transfer to the corrugated sheets of the structured packing:

$$Sh = 0.246 \quad (Sc\, Ga)^{0.33} E^{0.19} \tag{10}$$

It should be mentioned that in Eq. (10) the exponent 1/3 of the (Sc Ga) group had been adopted according to the model of Sigrist. With the present experimental results, the values of the exponents of (Sc Ga), indicated in Eqs. (8) and (9), give a better fit.

5. Concluding remarks

Gas holdup and liquid to wall mass transfer rates were measured simultaneously in a square bubble column with and without structured packing.

In presence of structured packing the measured gas holdups could be satisfactorily predicted by an expression obtained under similar conditions in a circular column. The validity of this expression is extended to higher fluid viscosities in this work.

In absence of the packing, no coincidence of gas holdup results for square and circular columns could be found. However, the appearance of slug flow with the most viscous solutions was common to both column shapes.

Single correlations describe liquid to wall mass transfer measured for the packed and unpacked columns, excluding in the latter case the results obtained with non-Newtonian fluids. For the last, the mass transfer data were correlated separately, since they correspond to a different flow regime.

The most remarkable finding is that, in contrast to what happens in heat exchangers with flowing fluids, the transfer rate to the container wall is almost the same with or without the structured packing.

Acknowledgement

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