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# Mass transfer to regular packings at low Reynolds numbers and under natural convection

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**Abstract**—Mass transfer coefficients for regular packings determined for low flow velocities show to be influenced by natural convection. In order to apply the coupling rule for forced and natural convection, mass transfer correlations for both pure regimes are required. Since no relationship for natural convection could be found in the literature, a systematic study of free convection in regular packings was undertaken. General correlations were developed and it was shown, that they also apply to static mixers. A simple model was formulated which describes natural convection mass transfer in structured packings by correlations obtained for inclined surfaces. The mass transfer results measured for low Reynolds numbers could be correlated successfully by the coupling rule and also by a method using a modified Reynolds number. 

## 1. INTRODUCTION

Structured packings are nowadays widely used in the process industry. Due to their high efficiency they are replacing random packings and trays in many equipments. Structured packings are not only used in thermal separation technology, but find application in many different fields, as in heat exchange, liquid fluidization, dust washing, static mixing, homogenization, in-line dispersion, etc. In recent years the use of regular packings as supports in catalytic chemical reactors [1] and in biofilm reactors [2] has been suggested. These devices, with the catalyst or the microorganisms deposited on the packing surface get benefit from the excellent macromixing characteristics and the high mass transfer efficiency of the structured packings.

Due to the important applications of regular packings, their behaviour under different operating conditions has been investigated. Mass transfer for structured packings in single phase [3], in fluidized beds [4, 5], in bubble columns [6] and in solid suspended bubble columns [7] has been studied experimentally.

When the microorganisms immobilized on the packing surface present a slow growth rate, a high residence time is needed in order to obtain a high bio-conversion. In other words, the bioreactors must operate at low liquid velocities. For this reason it is interesting to study the mass transfer behaviour of regular packings for low liquid flow rates and for the limiting case of absence of externally forced flow.

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## 2. EXPERIMENTAL

Mass transfer rates from the liquid to the surface of the packing were determined by the well-known limiting current technique [8].

The arrangement of equipment is shown in Fig. 1.

The experiments were performed using a square section column made of perspex (4 × 4 cm, 34 cm high) containing three to five packs of structured packing. The electrolyte was circulated through the column from a constant temperature reservoir, with superficial velocities varying from 0.016–4.36 cm/s. At the higher flow rates a centrifugal pump and flowmeters were used. In the lower range a constant head tank had to be used and the flowrate was calculated from the volume collected at the outlet of the column for a given period of time. By changing the lines, an inverted column could be used for downflow studies. Experimental uncertainty in the velocity measurement was estimated to be about 4%.

For the free convection measurements the column was placed in the thermostat in order to control the electrolyte temperature accurately.

For the mass transfer measurements by the limiting current method two of the metal sheets of the middle pack were connected to the external circuits through thin insulated wires. These electrically active plates, insulated from the neighboured sheets by epoxy coating, were used as the cathode in the electrochemical reduction of ferricyanide ions. The adjacent packs, with all sheets in electrical contact, were used as the anode. The measurement uncertainty of the convection mass transfer coefficient, due primarily to uncertainty in the titration of the electrolyte and in geometry measurements, was less than 5%.

Table 1 summarizes the characteristics of the reg-

## NOMENCLATURE

| | |
|---|---|
| $a$ | surface area per unit volume [$m^2\ m^{-3}$] |
| $a'$ | channel side [m] |
| $b$ | corrugation wavelength [m] |
| $c$ | electrolyte concentration [$kmol\ m^{-3}$] |
| $c_0$ | electrolyte concentration in bulk [$kmol\ m^{-3}$] |
| $c_p$ | specific heat [$kJ\ kg^{-1}\ K^{-1}$] |
| $d$ | disk diameter, sphere diameter [m] |
| $D$ | diffusivity [$m^2\ s^{-1}$] |
| $D'$ | width of corrugated plate [m] |
| $D_h$ | hydraulic diameter $= [a' \sin\alpha]/[1+\sin(\alpha/2)]$ [m] |
| $g$ | gravitational acceleration [$m\ s^{-2}$] |
| $Gr_d$ | Grashof number $= g\alpha^* c_0 d^3/v^2$ |
| $Gr_{L^*}$ | Grashof number $= g\alpha^* c_0 L^{*3}/v^2$ |
| $Gr_Z$ | Grashof number $= g\alpha^* c_0 Z^3/v^2$ |
| $h$ | convective heat transfer coefficient [$W\ m^{-2}\ K^{-1}$] |
| $j$ | $j$-factor for mass transfer $= Sc^{2/3}\ k/v$ |
| $k$ | mass transfer coefficient [$m\ s^{-1}$] |
| $k'$ | thermal conductivity [$W\ m^{-1}\ K^{-1}$] |
| $L^*$ | characteristic length of regular packing, equation (5) [m] |
| $Nu_d$ | Nusselt number $= hd/k'$ |
| $Pr$ | Prandtl number $= \mu/c_p k'$ |
| $Re$ | Reynolds number $= vD_h/v$ |
| $Re_d$ | Reynolds number $= v_s d/v$ |
| $Re_Z^*$ | modified Reynolds number, equation (11) |
| $Re_Z$ | Reynolds number $= v_s Z/v$ |
| $Sc$ | Schmidt number $= v/D$ |
| $Sh_d$ | Sherwood number $= kd/D$ |
| $Sh_{L^*}$ | Sherwood number $= kL^*/D$ |
| $Sh_{ZM}$ | Sherwood number for mixed convection, equation (2) |
| $Sh_Z$ | Sherwood number $= kZ/D$ |
| $Sh_\gamma$ | mean Sherwood number for inclined surfaces, equation (6) |
| $u_m$ | maximum flow velocity in natural convection boundary layer, equation (13) [$m\ s^{-1}$] |
| $v$ | interstitial velocity [$m\ s^{-1}$] |
| $v_s$ | superficial velocity, free-stream velocity [$m\ s^{-1}$] |
| $X$ | liquid layer depth between packing elements [m] |
| $z$ | distance from leading edge [m] |
| $Z$ | length of flat or corrugated plate or packing element [m]. |

Greek symbols

| | |
|---|---|
| $\alpha$ | folding angle of furrow [°] |
| $\alpha^*$ | densification coefficient $= \Delta\rho/\Delta c\rho$ [$m^3\ kmol^{-1}$] |
| $\beta$ | corrugation inclination angle respect to vertical [°] |
| $\gamma$ | surface inclination angle respect to vertical [°] |
| $\varepsilon$ | void fraction |
| $\lambda$ | free convection weighting factor in modified Reynolds number |
| $\mu$ | dynamic viscosity [mPa s] |
| $\nu$ | kinematic viscosity [$m^2\ s^{-1}$] |
| $\rho$ | density [$kg\ m^{-3}$]. |

Table 1. Characteristics of structured packings

| Packing | $\alpha$ [°] | $\beta$ [°] | $\gamma$ [°] | $a$ [$cm^{-1}$] | $a'$ [cm] | $b$ [cm] | $D_h$ [cm] | $L^*$ [cm] | $Z$ [cm] |
|---|---|---|---|---|---|---|---|---|---|
| A | 97.0 | 30 | 20.9 | 8.23 | 0.567 | 0.85 | 0.308 | 2.916 | 4.00 |
| B | 95.0 | 30 | 21.5 | 8.20 | 0.572 | 0.85 | 0.314 | 3.240 | 5.00 |
| C | 106.5 | 40 | 26.7 | 8.24 | 0.562 | 0.90 | 0.286 | 2.800 | 4.00 |
| D | 108.9 | 45 | 30.2 | 8.20 | 0.614 | 1.00 | 0.307 | 2.545 | 4.00 |
| E | 108.9 | 45 | 30.2 | 8.00 | 0.614 | 1.00 | 0.307 | 2.545 | 3.30 |
| F | 82.4 | 45 | 30.2 | 5.68 | 0.532 | 0.70 | 0.297 | 3.174 | 4.42 |
| G | 106.5 | 50 | 35.5 | 8.37 | 0.562 | 0.90 | 0.286 | 2.800 | 4.00 |
| H | 97.0 | 60 | 48.8 | 8.05 | 0.567 | 0.85 | 0.308 | 2.916 | 4.00 |

Average void fraction for all packings $\varepsilon = 0.88$.
Average thickness of metal sheet e = 0.025 cm.
For definition of geometric parameters see Fig. 4.

ular packings employed. The forced convection experiments were carried out with only one type of structured packing, namely the one mostly used in practice, which has a corrugation angle respect to flow direction of 45°.

During the experimental study of natural con-

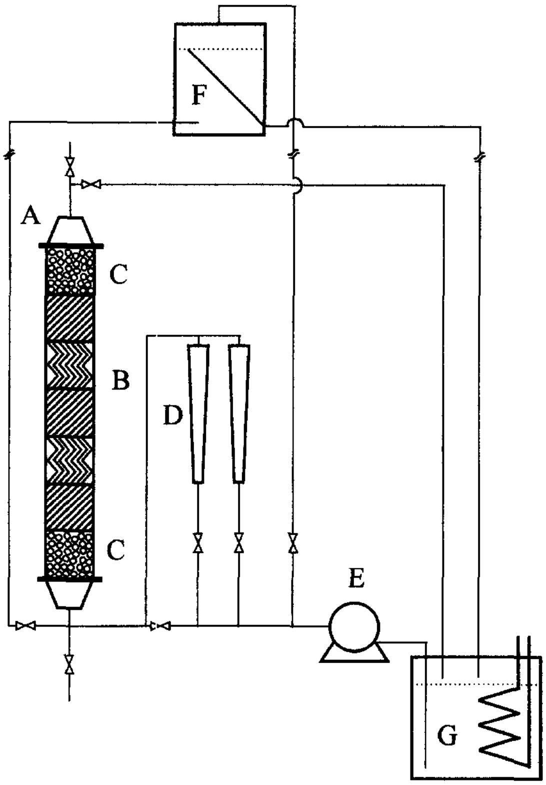


Fig. 1. Experimental equipment. A: column of square section, B: structured packings, C: calming section, D: flowmeters, E: centrifugal pump, F: constant head tank, G: storage tank with thermostat.

vection additional configurations were investigated as will be described.

The range of experimental variables and derived quantities are summarized in Table 2.

### 3. FORCED FLOW EXPERIMENTS

Mass transfer coefficients obtained with solutions S1 to S4 are shown in Fig. 2 as a function of the interstitial velocity. For both, upward and downward flow direction almost the same values were obtained. Therefore in the following no distinction will be made between both configurations. On the $y$-axis the mass transfer coefficients obtained for solutions S2, S3 and S4 without fluid circulation are also shown. Free convection results for solution S1 were not reproducible, this is characteristic of the free convection with small driving force [8]. As can be seen, at the lowest flow rates mass transfer coefficients approach the free convection values.

In Fig. 3 all the forced flow data of Fig. 2, irrespective of the flow direction, are presented in dimensionless form together with an empirical correlation previously developed for Reynolds numbers ranging from 10–135, in the domain of laminar forced convection [3]:

$$j = 0.704\,Re^{-0.526}. \qquad (1)$$

$Re$ is based on the hydraulic diameter of the packing as the characteristic length and the velocity is the interstitial flow velocity. As the Reynolds number decreases, $j$-factors diverge from the correlation of Colazo *et al.* [3] for almost all solutions yielding higher values as predicted. Buoyancy forces seem to affect mass transfer.

The attempt of improving the correlation of experimental data obtained in the low Reynolds number range—for instance by the now common coupling rule suggested by Churchill [9]—led to the need of investigating natural convection in structured packings in more detail. The mixed convection correlation of the Churchill type combines independently a forced convection term and a natural convection term derived from the corresponding correlations, each raised to the same power:

$$Sh^{m}_{ZM} = Sh^{m}_{ZFC} \pm Sh^{m}_{ZNC}. \qquad (2)$$

The exponent m characterizes the coupling of both convection mechanisms; the plus and minus signs refer to assisting and opposing mechanisms, respec-

Table 2. Electrolytic solution characteristics at 25°C and range of derived quantities

| Solution | $[Fe(CN)_6^{3-}]$ [kmol m$^{-3}$] | [NaOH] [kmol m$^{-3}$] | $\rho \times 10^{-3}$ [kg m$^{-3}$] | $\mu$ [mPa s] | $D \times 10^{10}$ [m$^2$ s$^{-1}$] |
|---|---|---|---|---|---|
| S1 | 0.001 | (*) | 1.060 | 1.167 | 5.59 |
| S2 | 0.010 | (*) | 1.060 | 1.200 | 5.51 |
| S3 | 0.025 | 0.5 | 1.030 | 0.950 | 7.84 |
| S4 | 0.050 | 0.5 | 1.040 | 0.970 | 7.68 |
| S5 | 0.025 | 2.0 | 1.080 | 1.355 | 5.48 |
| S6 | 0.050 | 2.0 | 1.095 | 1.394 | 5.34 |
| S7 | 0.100 | 2.0 | 1.117 | 1.494 | 4.98 |
| S8 | 0.200 | 2.0 | 1.154 | 1.642 | 4.54 |

For all solutions a value of $\alpha^* = 0.025$ m$^3$ kmol$^{-1}$ was adopted.
Schmidt numbers ranged from 1176 to 3135.
Grashof numbers $Gr_z$ varied from $1.29 \times 10^4$ to $1.55 \times 10^6$.
(*) The supporting electrolyte was a buffer of $Na_2CO_3/NaHCO_3$ 0.4 M.

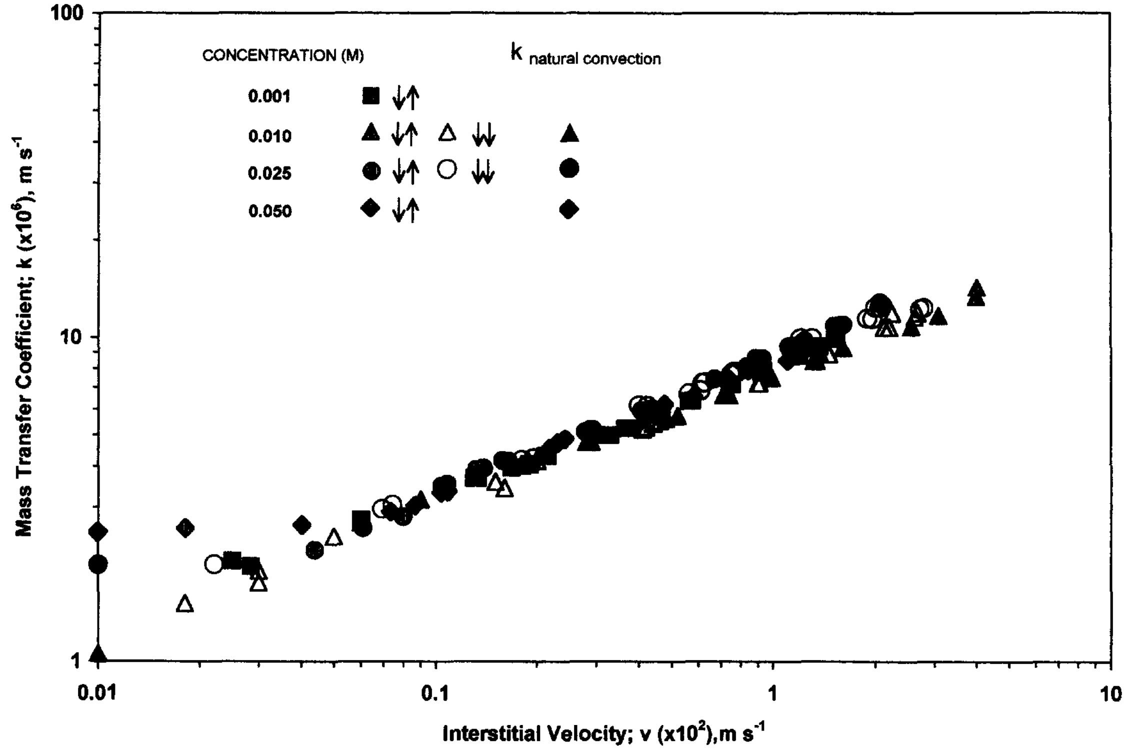


Fig. 2. Mass transfer coefficients as function of interstitial velocity in regular packing (Packing F). (Arrows indicate directions of natural and forced convection flow.)

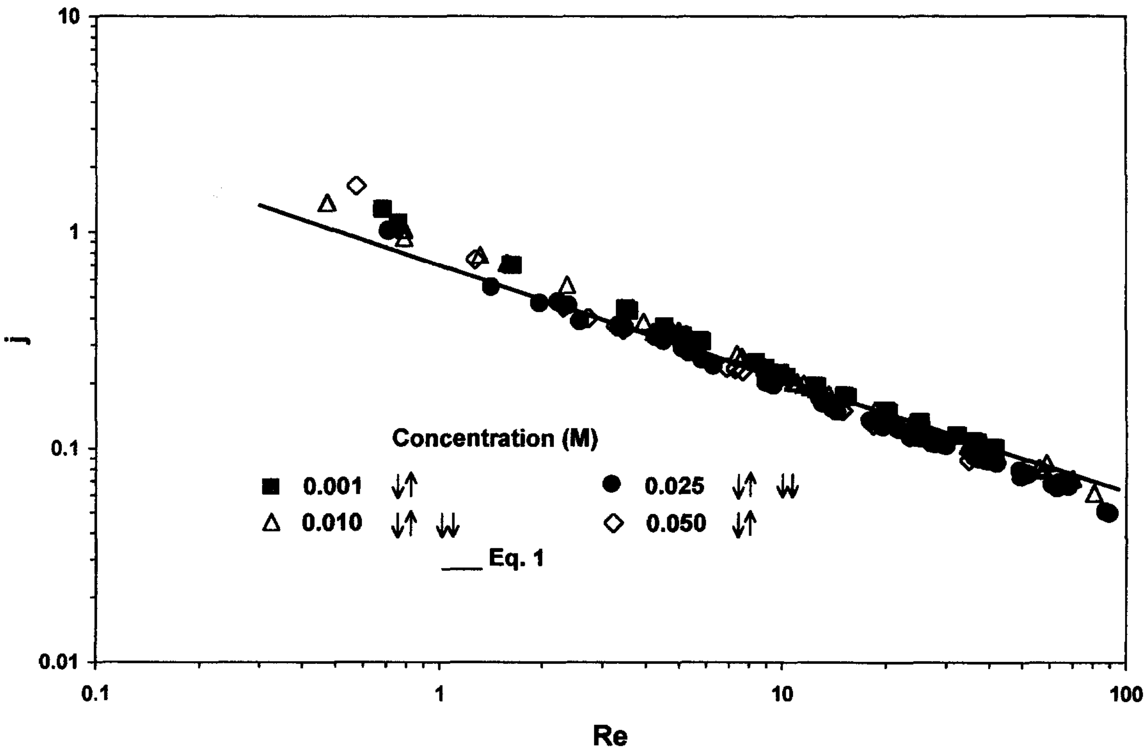


Fig. 3. General mass transfer correlation.

tively. The forced convection component may be obtained from the correlation developed by Colazo *et al.* [3], but, to the best knowledge of the authors, no correlation is available in the literature which could predict the natural convection component. Hence, a major objective of the present investigation has been to develop such a correlation for the particular geometry of structured packings.

## 4. NATURAL CONVECTION EXPERIMENTS

The experiments were performed using solutions S5 to S8. The physical properties of these solutions are well known and the migration contribution to mass transfer, even for the highest concentration of reactive ions, is negligible [10]. The apparent characteristic densification coefficient defined by Hiraoka *et al.* [11] was used to relate the density difference built up in the boundary layer with the corresponding concentration gradient. A constant value of $\alpha^* = 0.025\ m^3/kmol$ was adopted for all solutions, since it was shown elsewhere [11] that the characteristic densification coefficient only varies slightly over the concentration range employed.

Preceding the main experiments, mass transfer coefficients for natural convection from a vertical flat plate having the same length as the corrugated plates were determined for comparison purposes. The results are adequately correlated by the well known correlation:

$$Sh_Z = 0.67(Sc\,Gr_Z)^{1/4}. \tag{3}$$

The validity of the experimental technique and the accuracy of the physical properties is thus confirmed.

Figure 4 shows the different arrangements of corrugated sheets investigated.

System I consists of a single corrugated plate acting as the cathode. It was placed in the centre of the column with two packs of structured packing located above and below, respectively. These packs, without any insulation, were used as the counterelectrode. The single plate, surrounded by electrolyte solution only, allows natural convection to develop freely. No channels confine the boundary layer and no interaction with neighboured channel flows exists. The corrugations may be compared to V-groove roughness elements of a planar electrode which act as surface promoters. Since the aim of the present investigation is the study of natural convection mass transfer in structured packings, results obtained with system I will only be used to check whether the electrodes were working properly and for comparison purposes.

In system II the same corrugated plate was placed in the middle of an electrically inactive pack, which was arranged between the anodic packs. Thus, both surfaces of the cathode were facing inert corrugated plates.

In system III two adjacent corrugated plates of the middle pack were connected as cathodes. The outer surfaces of these sheets were isolated with lacquer to make them inactive; thus, two active surfaces faced each other. The last two systems correspond precisely to a structured packing. As usual, subsequent elements were rotated by 90°.

Experiments for these three geometries were carried out and repeated many times, for all the structured packings described in Table 1. They mainly differ in the inclination angle $\beta$ of the triangular-shaped furrows. The angled corrugations of adjacent sheets are reversed with respect to each other forming a series of connected intersections of channels. Interaction of the criss-crossing streams, if there is any, may depend on this inclination angle.

The experimental results obtained with the three systems, for all inclination angles and all electrolyte concentrations investigated, are depicted in Fig. 5, where the mean mass transfer coefficient, $\bar{k}$, is plotted as a function of the inclination angle. The $\bar{k}$-values obtained with the flat plate are also shown. As expected, mass transfer rates with corrugated sheets are always higher than with flat plates. The influence of the corrugation angle is most pronounced for the single corrugated plate, followed by system III (with two active surfaces facing each other) and levels down for system II (with the active sheet neighboured by inert corrugated plates). In all three cases the mass transfer coefficient goes through a maximum in the range of 45–50° of corrugation inclination, $\beta$, in accordance with similar findings described in the literature [12, 13].

Focke *et al.* [12] who studied the effect of corrugation angle on the performance of heat exchangers, found that the driving force inducing swirling motions in the furrows is maximum when $\beta = 45°$. Spekuljak and Andrada [13] who investigated the influence of geometrical variables on the performance of static mixers, concluded that the best mixing properties are obtained for corrugation angles in the 40° range.

Comparing the behaviour of systems II and III, it can be seen that for $\beta = 30$ and 60°, mass transfer coefficients are practically identical. The same happens for all angles at the lowest electrolyte concentration, which only produces weak free convection. Differences begin to appear for the angles of 45 and 50°. These differences increase with increasing solution concentration and extend to the lower inclination angles for the highest electrolyte concentration.

## 5. CORRELATION OF NATURAL CONVECTION DATA

Experimental results were correlated in the classic dimensionless form:

$$Sh_Z = A_i(Sc\,Gr_Z)^{B_i} \tag{4}$$

using the height $Z$ of the plate or pack as characteristic length. Table 3 lists the values of the parameters $A_i$ and $B_i$ which best fit the experimental data for each

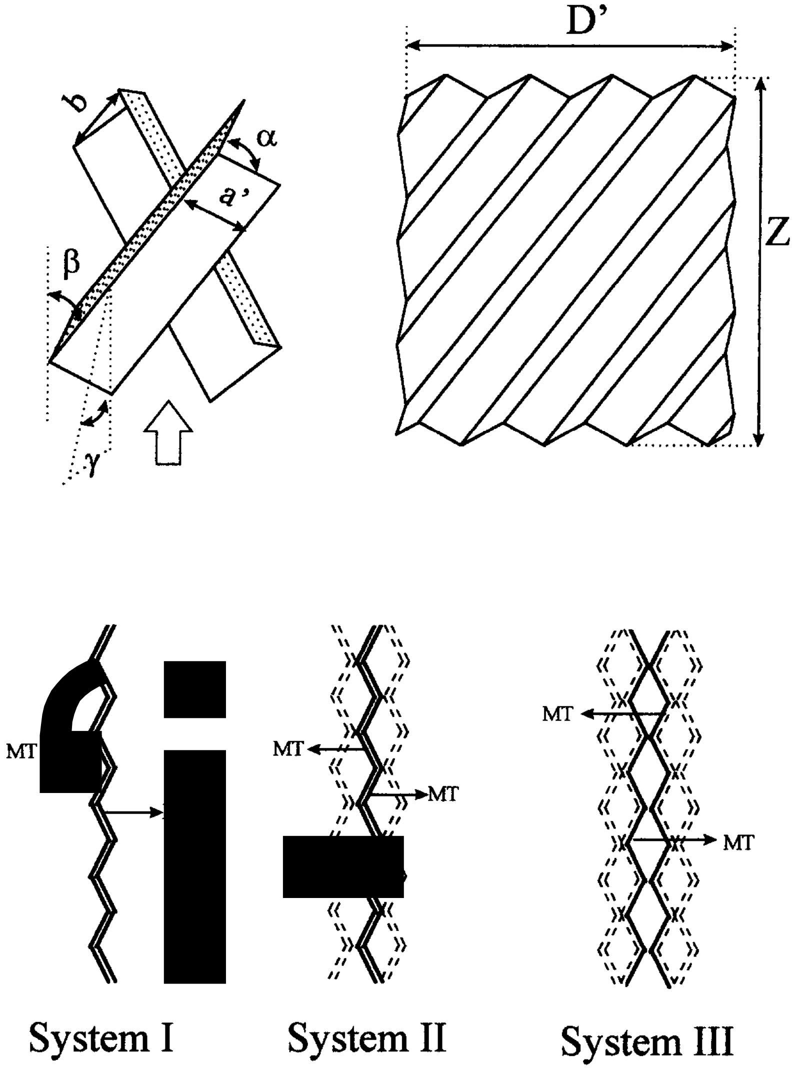


Fig. 4. Definition of geometric parameters and arrangements of corrugated sheets.

system and each inclination angle. In Fig. 6 the correlations obtained for system II are plotted as an example.

Examination of Table 3 shows that for the single corrugated plate there is an important variation of the exponent of $Sc\,Gr_Z$ and the numerical coefficient $A_i$, with the inclination angle. The values of the exponent indicate that for the lowest and for the highest inclination angle studied, laminar flow conditions exist, meanwhile for the other angles, transition to turbulent flow is inferred.

For system II, with the active surfaces facing inert ones, the differences from angle to angle are less pronounced. On the other hand, for system III, for all inclination angles, except $\beta = 30°$, the exponent is close to 1/3, which applies for turbulent flow. This may be explained by the fact that natural convection streams are generated in opposed channels, since both surfaces are active, and interaction of crossing flows may occur producing swirl motion.

Although mass transfer coefficients may vary with the inclination angle by some percents (the maximum difference between mean $k$-values is 14.4%, corresponding to system III, the highest electrolyte concentration and angles of 50 and 30°); it can be seen that in general differences are small. So, for practical purposes, all the experimental data—independently of the inclination angle—were subjected to statistical analysis giving general correlations for systems II and III. The results are shown in Table 4; the equations that fit the data are seen to be quite similar for both systems.

In an attempt of taking into account the corrugation inclination angle, the characteristic length $L^*$, derived for structured packings [14],

$$L^* = D'Z/(D'\cos\beta + Z\sin\beta) \tag{5}$$

was introduced rather than the pack height. The general correlations which are derived, are included in

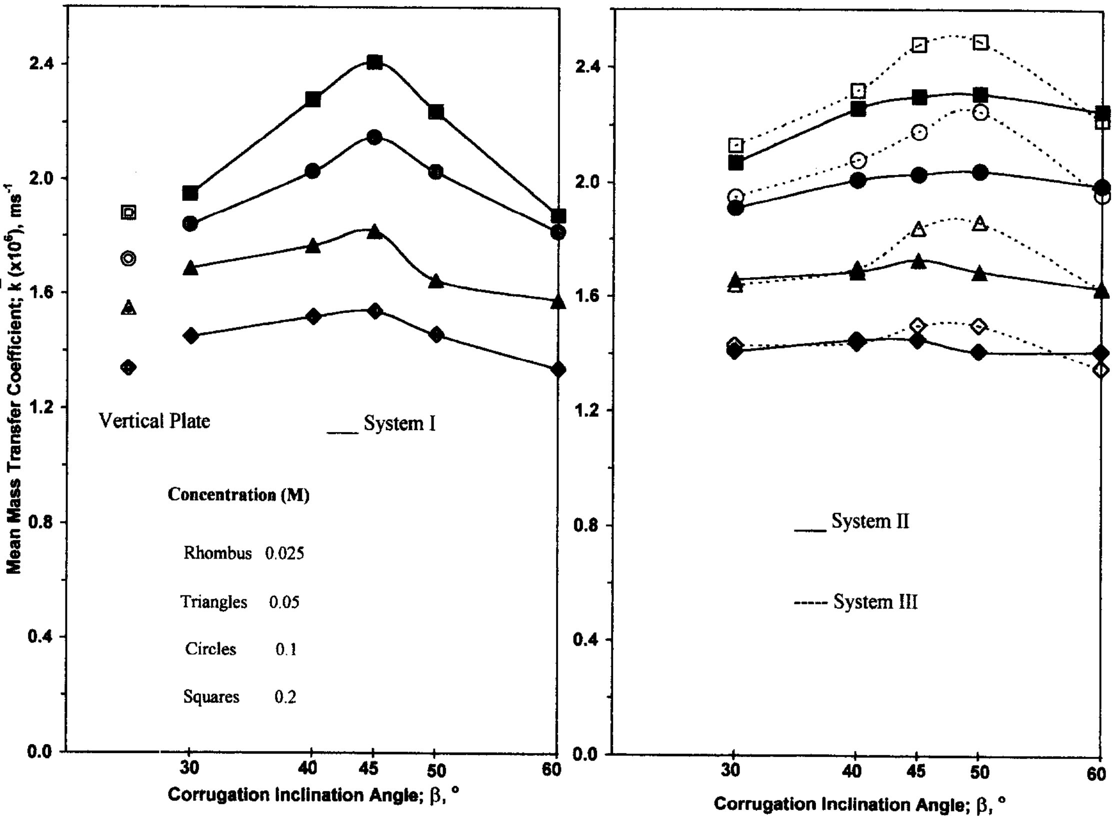


Fig. 5. Natural convection mass transfer coefficients for flat and corrugated plates and for packings of corrugated plates ($Z = 4$ cm). Influence of corrugation inclination angle on natural convection mass transfer.

Table 3. Numerical values of $A_i$ and $B_i$ in equation (4) for each corrugation inclination angle

| | System I | | System II | | System III | |
|---|---|---|---|---|---|---|
| $\beta$ | $A_1$ | $B_1$ | $A_2$ | $B_2$ | $A_3$ | $B_3$ |
| 30 | 0.926 | 0.235 | 0.447 | 0.270 | 0.361 | 0.281 |
| 40 | 0.400 | 0.279 | 0.248 | 0.300 | 0.172 | 0.318 |
| 45 | 0.260 | 0.301 | 0.225 | 0.305 | 0.159 | 0.324 |
| 50 | 0.258 | 0.298 | 0.153 | 0.323 | 0.147 | 0.328 |
| 60 | 0.626 | 0.251 | 0.182 | 0.314 | 0.144 | 0.324 |

Table 4 and are seen to be very similar to those obtained with $Z$. Also, the similitude of the equations in terms of $L^*$ describing the mass transfer behaviour of systems II and III is remarkable. Either of these characteristic dimensions may be recommended for design calculation purposes.

## 6. COMPARISON WITH PREVIOUS MASS TRANSFER STUDIES

Since a structured packing is composed of vertically oriented corrugated sheets with triangular-shaped inclined furrows, it may be modelled as an assemblage of inclined upward and downward facing strips.

In the past, natural convection at inclined surfaces has been examined by several authors [15–19] and two mechanisms of mass transfer were described: attached boundary layer flow conditions and separation of the initially attached boundary layer. In the present investigation, where the cathodic reduction of ferricyanide was measured, the free convection flow is directed downwards, so that the first mechanism appears at the upwards facing surfaces and the second one may appear at the downwards facing surfaces, depending on the inclination angle and the electrolyte concentration. However, since both kinds of surfaces have common edges, mass transfer rates at the different surfaces will not be additive because of significant

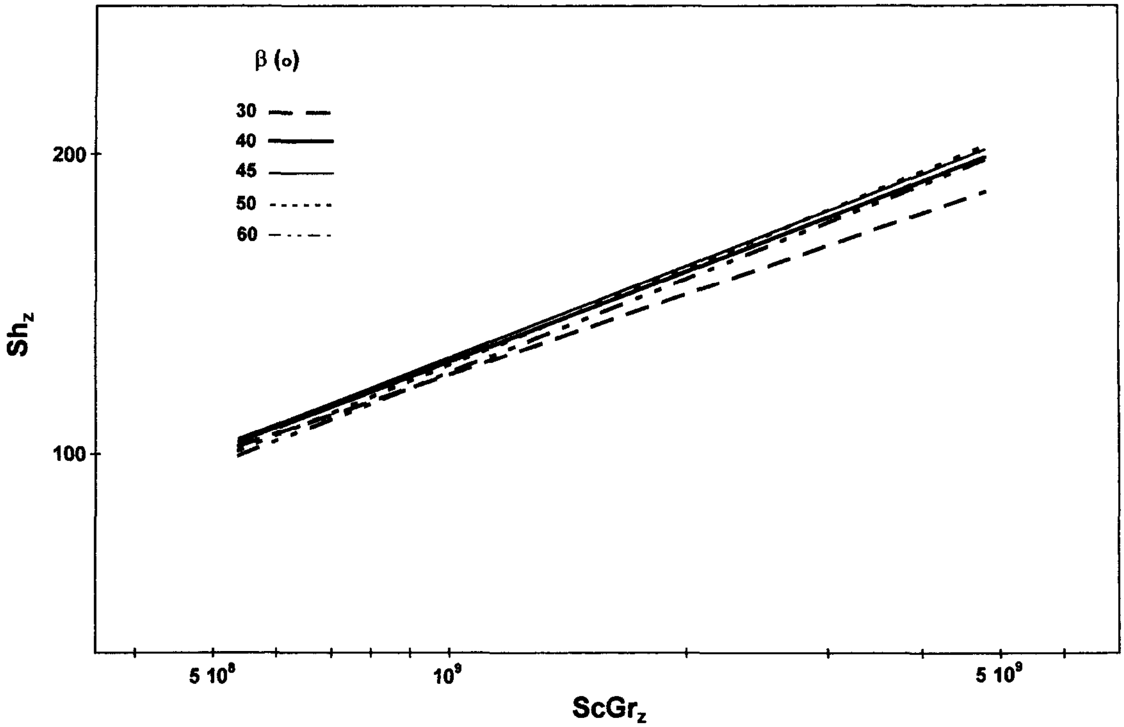


Fig. 6. Natural convection mass transfer correlations for structured packings with different corrugation inclination angles (all packings; active surfaces facing inert ones).

Table 4. General correlations for natural convection mass transfer in structured packings using different characteristic lengths; $Sh_Z = A_i(Sc\,Gr_Z)^{B_i}$ and $Sh_{L^*} = A_j(Sc\,Gr_{L^*})^{B_j}$

| | System II $N = 51$ | | | System III $N = 80$ | | |
|---|---|---|---|---|---|---|
| | $A_2$ | $B_2$ | $\delta$ (%) | $A_3$ | $B_3$ | $\delta$ (%) |
| $f(Z)$ | 0.252 | 0.299 | 2.3 | 0.227 | 0.305 | 4.5 |
| $f(L^*)$ | 0.246 | 0.298 | 2.4 | 0.247 | 0.300 | 4.24 |

$N$ = number of experimental data.
$\delta$ = mean deviation.

$$\delta = \frac{1}{N}\sum_1^N \frac{|Sh\,\mathrm{calc} - Sh\,\mathrm{exp}|}{Sh\,\mathrm{exp}}.$$

interactions and a summation approach is not suitable. Instead, a geometric mean approach is tested:

$$Sh_\gamma = (Sh_1\,Sh_2)^{1/2}. \quad (6)$$

Here $Sh_1$ and $Sh_2$ refer to up and downfacing inclined surfaces, respectively. The mean length of these surfaces, constituting the sides of the furrows, is given by $L^*$ and the inclination angle of the surface respect to the vertical is calculated by the formula derived by Spekuljak [14]:

$$\gamma = \tan^{-1}\left[\frac{\sin\beta\cos(\alpha/2)}{\cos\beta}\right]. \quad (7)$$

Among the correlations given in the literature, those of Patrick et al. [19] were selected to predict mass transfer rates for up and downfacing surfaces. These correlations are:

$$Sh_1 = 0.68(Sc\,Gr\cos\gamma)^{1/4} \quad (8)$$

and

$$Sh_2 = c'(Sc\,Gr\cos\gamma)^{1/3} \quad (9)$$

with

$$c' = 0.063 + 0.002314\gamma.$$

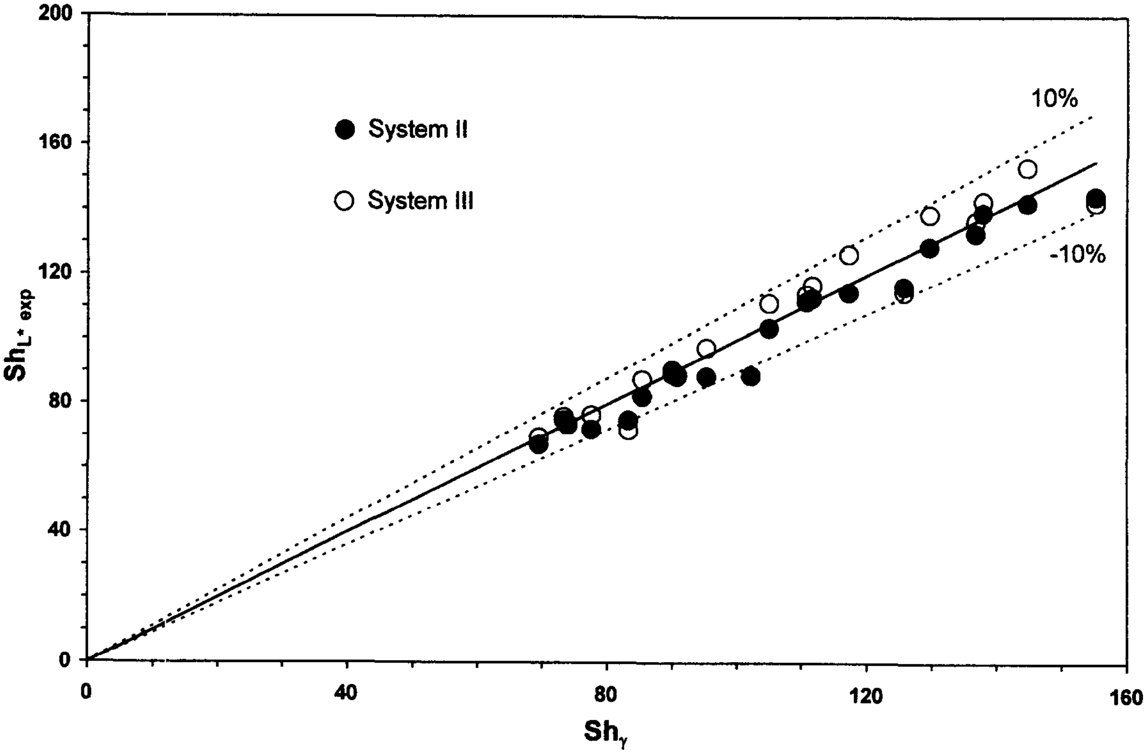


Fig. 7. Comparison between experimental Sherwood numbers and Sherwood numbers calculated with the aid of mass transfer correlations for inclined surfaces (free convection).

In Fig. 7 the experimental *Sh*-values for structured packings (systems II and III) are plotted against the mean *Sh* number obtained by applying literature correlations for inclined surfaces and using equation (6). The calculated and experimental values show a difference greater than 10% only in a few cases. In view of the fact that the mass transfer system is highly complex, predictions are quite satisfactory and the procedure presented above can also be used to obtain natural convection mass transfer coefficients in structured packings.

## 7. FURTHER STUDIES OF NATURAL CONVECTION IN STRUCTURED PACKINGS

Some exploratory experiments were also carried out with the packing elements placed in the column in the usual manner, i.e. rotating one pack respect to the next one by 90°, but with liquid layers of different depth between subsequent elements: a geometry often used in practice in static mixing. Separations between packs were varied up to 5 cm beginning with the elements stacked close together.

Some of the results are depicted in Fig. 8. No significant variation of the mass transfer coefficient with the distance between consecutive packs can be observed. It can be concluded that there are no appreciable end effects affecting mass transfer inside the element of structured packing. Therefore, mass transfer correlations derived for the stacked packings can also be applied to static mixers.

Furthermore, separation of packs allowed visualization by a dark field method [20] of the natural convection streams leaving the channels of the pack. A suitable fraction of lycopodium particles was suspended in the electrolyte and the particles accompanying the stream were illuminated by a laser beam spread into a vertical plane beneath the working pack. Figure 9 shows the time exposure photograph of the flow pattern obtained with system III, $\beta = 30°$, $c_0 = 0.025$ kmol/m$^3$. Streams leaving the inclined channels pertaining to one corrugated plate become mixed with the streams coming in reversed direction from the adjacent sheet producing turbulent motion, thus mixing the fluid. The streams themselves seem to be in laminar motion.

## 8. CORRELATION OF COMBINED CONVECTION DATA

As has been mentioned before, the mass transfer correlations which apply to the combined natural and forced convection are based on the power sum of the individual Sherwood numbers corresponding to the two limiting cases of pure forced and pure free convection [equation (2)]. The Sherwood number where forced convection exists alone can be estimated from

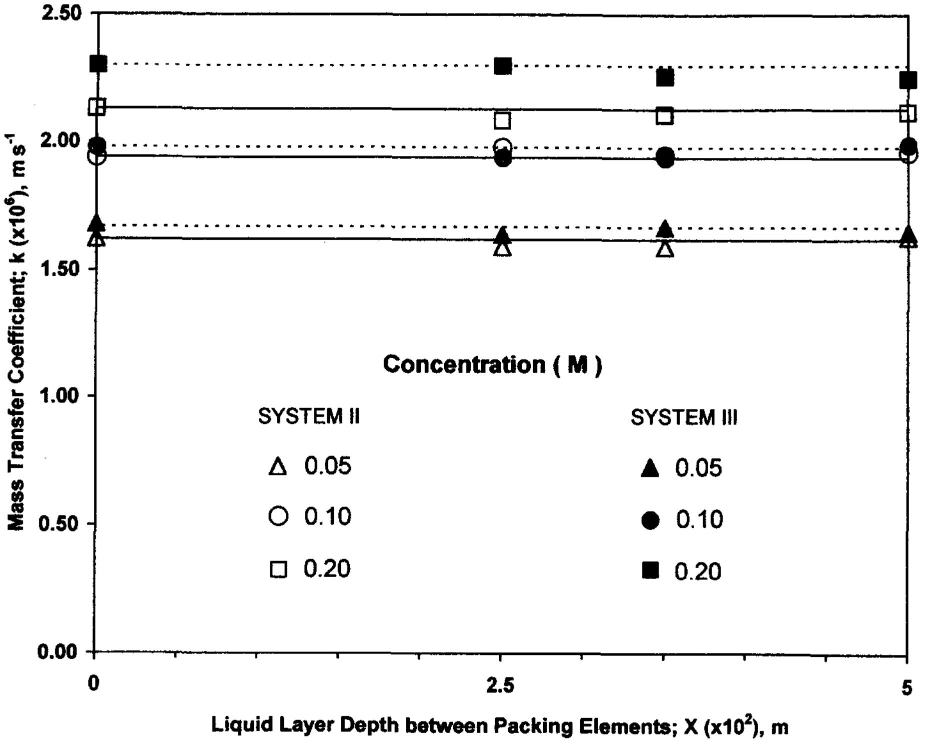


Fig. 8. Natural convection mass transfer coefficients for structured packings and static mixers. Influence of liquid layer depth between packing elements (Packing B).

Fig. 9. Flow pattern of natural convection streams leaving a structured packing element (Packing A, solution S5, exposure time 15 s).

the $k$-values given by equation (1) which holds for $Re \leqslant 135$.

For the estimation of the Sherwood number value corresponding to pure natural convection the general relationship (Table 4)

$$Sh_Z = 0.252(Sc\,Gr_Z)^{0.299} \qquad (10)$$

was selected, since the forced flow experiments were carried out with the geometry corresponding to active surfaces facing inert ones. Nevertheless, the general correlation derived for active surfaces facing each other, would give almost the same value due to the low electrolyte concentrations used in the forced flow experiments.

It should be mentioned that the $k$-values shown in Fig. 2 obtained for no net circulation in the forced flow column, are in good agreement with those estimated from equation (10).

Since the boundary between combined and pure forced convection is not sharp (Fig. 3) the data pertaining to the domain of $Re < 5.5$, were chosen to be checked by the coupling rule. As mentioned, for both assisting and opposing flows no differences in the mass transfer rates were detected, possibly due to the swirling motion within the channels, which tends to augment mass transfer coefficients in every case. In a flow visualization study in ducts with corrugated plates [21] spiralling in the flow along a furrow was found due to the interaction between the criss-crossing flows. On the other hand, as described by Jorné [22], separation of boundary, flow reversal and internal circulation may occur even at vertical electrodes with opposing flows. Therefore the combining rule was attempted indistinctly for both, upflow and downflow configurations and in view of the positive deviation of the $j$-factors, in equation (2) only the plus sign was applied.

The correlating power m in the interpolation formula was varied from 2–6. It was found that the quadratic mean error of $Sh_{ZM}$ decreased from 18.7 to 11.8% for m increasing from 2–4, then remaining almost constant. Figure 10 shows the results obtained with m = 4 in comparison with the experimental data. Only a few data present differences greater than 15% between experimental and predicted values.

Inspection of Fig. 10 shows that the power sum of individual Sherwood numbers with m = 4 can be used in order to evaluate mixed convection mass transfer to structured packings with sufficient accuracy for practical purposes.

As commented in the literature [23], aside from the combining rule mentioned above, another method, which uses a modified Reynolds number, has been remarkably successful in describing combined forced and natural convection. This modified Reynolds number incorporates a characteristic buoyancy-induced velocity added to the imposed free-stream velocity:

$$Re_d^* = Re_d + \lambda(Gr_d/2)^{1/2} \quad \text{or} \quad Re_Z^* = Re_Z + \lambda(Gr_Z/2)^{1/2}. \tag{11}$$

The method using the modified Reynolds number has the advantage of representing data for forced, mixed and natural convection by a single curve, since $Re_Z^*$ reduces to the asymptotes of pure natural and pure forced convection. The only difficulty consists in the correct choice of the weighting factor $\lambda$.

Kobus and Wedekind [23], who studied combined forced and natural convection heat transfer from circular disks to air, obtained:

$$\lambda = 0.723/\{(20/21)+Pr\}^{1/2}$$

$$\lambda = 0.558 \quad \text{for air at } 21^\circ\text{C}. \tag{12}$$

In their analysis they used the maximum velocity in the free convection boundary layer along the disk centreline as characteristic velocity to be added to the imposed free-stream velocity. The same model was tried in the present investigation. It is noted that the geometry of the structured packing under study, as well as the flow pattern in the furrows are very complex and the only aim is to obtain an approximate value of $\lambda$. Using the expression for the maximum velocity in the natural convection boundary parallel to a vertical flat electrode given by Ibl and Müller [20] with

$$u_m = K(g\alpha^* c_0 Dz/\nu)^{1/2} \tag{13}$$

and $K = 0.77$ as derived by Wilke *et al.* [24] for limiting rates of electrolysis at vertical plates and averaging over the electrode length $Z$ yields, after appropriate rearrangement:

$$\lambda = 0.726/Sc^{1/2}. \tag{14}$$

This relationship is almost identical with equation (12) since in the high Schmidt number range, usual in electrolyte solutions, the term 20/21 can be neglected. Thus, for the present research, the mean value of the weighting factor for the natural convection contribution is about 0.02.

In Fig. 11 all the experimental data of the present work, including the pure free convection data, are plotted as function of the modified Reynolds number, adopting as a first approximation the value of $\lambda = 0.02$. Also shown are the relationships developed by Colazo *et al.* [3] for forced convection mass transfer in structured packings, which are:

$$j = 0.704 Re^{-0.526} \quad Re \leqslant 135$$

$$j = 0.358 Re^{-0.388} \quad Re > 135. \tag{15}$$

Both equations (obtained with packing $F$) were suitably transformed to where the packing height $Z$ was the characteristic length instead of the hydraulic diameter of the packing and the superficial flow velocity replaced the interstitial velocity. They are depicted in Fig. 11 in the form $Sh/Sc^{1/3}$ vs $Re_Z^*$, bearing in mind that $Re_Z^*$ reduces to $Re_Z$ at the high end of the modified Reynolds numbers.

The pure natural convection correlation, equation (10), was also rearranged to be plotted against $Re_Z^*$ in the same graph:

$$\frac{Sh_Z}{Sc^{1/3}} = 0.252\, Sc^{-0.034}\left[2\left(\frac{Re_Z^*}{\lambda}\right)^2\right]^{0.299}. \tag{16}$$

For a mean Schmidt number of 2100 corresponding to the current research on natural convection, the relationship that appears at the low end of Fig. 11 is derived.

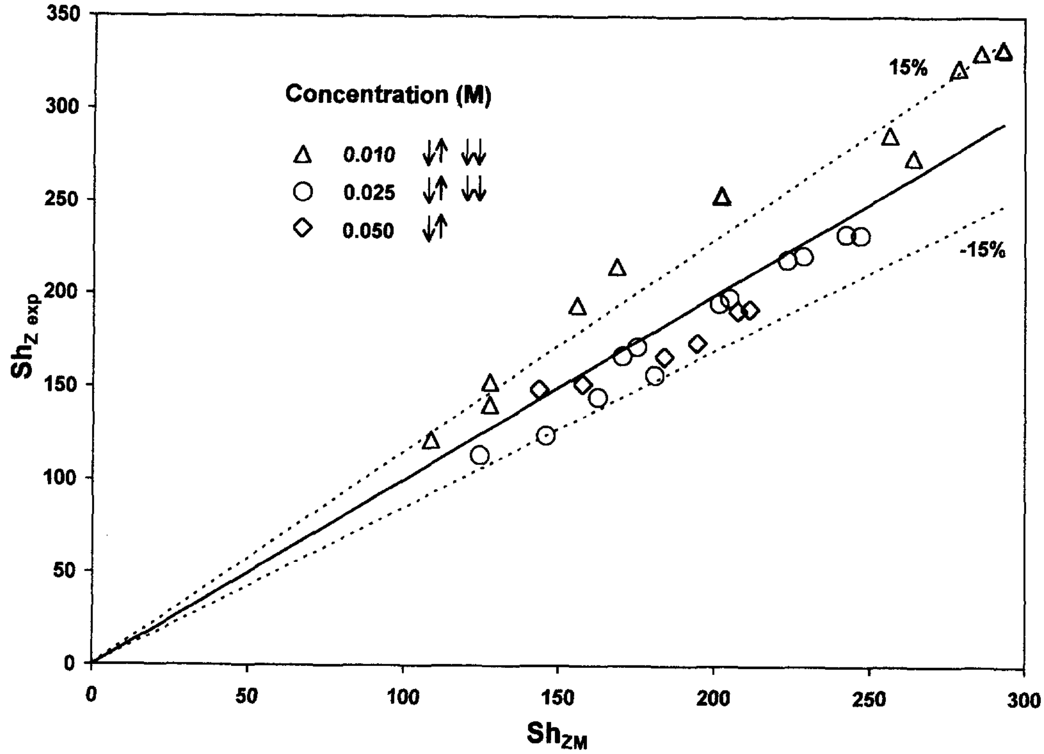


Fig. 10. Comparison of experimental Sherwood numbers and Sherwood numbers obtained by applying the coupling rule for combined forced and free convection.

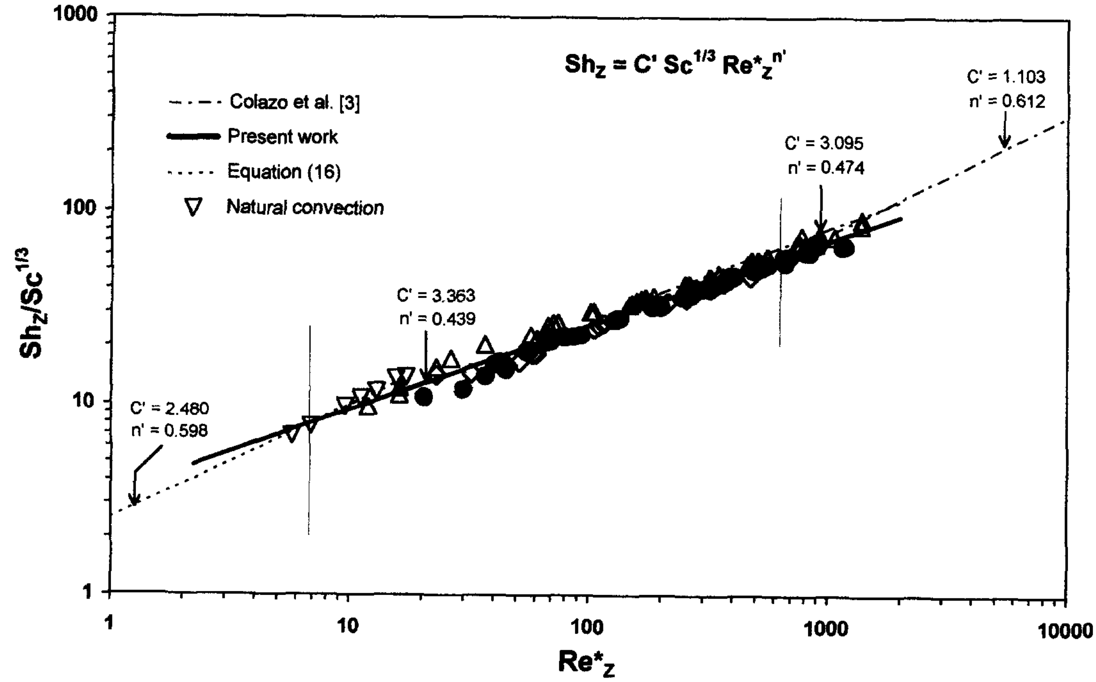


Fig. 11. Experimental results for natural, combined and forced convection flows in structured packings using a modified Reynolds number. Symbols as in Fig. 3.

In Fig. 11 it is apparent that the use of the modified Reynolds number allows to describe convection mass transfer to a regular packing by the general correlation

$$Sh_Z = C' Sc^{1/3} Re^{*n'}_Z \quad (17)$$

independent of the mode of convection. The values of $C'$ and $n'$ for the different domains are given in Fig. 11. There are no sharp limits between regions corresponding to pure free and combined convection or to combined and pure forced convection because of overlapping of data. Correlating lines intersect at $Re^*_Z = 6.8$ in the former case and at $Re^*_Z = 629$ in the latter case.

Figure 12 shows the correlating curve of the present work along with results of a study of mass transfer to one of the spheres in a packed bed of spheres at $Sc \cong 1600$, reported by Karabelas *et al.* [25]. The regions of pure natural, pure forced and combined convection are covered. As can be seen, the data of Karabelas *et al.* are in satisfactorily good agreement with the results of the present investigation, thereby lending support to the choice of the weighting factor $\lambda$.

The empirical correlations derived by Kobus and Wedekind [23] for forced, combined and natural convection heat transfer from vertical disks to air are also depicted together with the values of $C$ and $n$ to be used in the general heat transfer correlation

$$Nu_d = C\, Pr^{1/3} Re^{*n}_d. \quad (18)$$

Although, both continuous curves in Fig. 12 correspond to very different geometries, distinct characteristic lengths and $Pr$ or $Sc$ number values differing by a factor 1000, almost the same dependence on $Re^*$ is found in the mixed convection and pure forced convection domains. An exponent $n$ as low as that found by Kobus and Wedekind in their pure convection experiments is unknown in mass transfer correlations applying to natural convection mass transfer in liquids. However the use of a modified Reynolds number, as recommended by Kobus and Wedekind, shows that mass transfer rates in regular packings obey correlations of the same type over the whole range investigated, irrespectively of the transport mechanism involved. The correlation valid at low forced-flow rates

$$Sh_Z = 3.363\, Sc^{1/3} Re^{*0.439}_Z \quad (19)$$

(with a mean deviation of 7.2%), will predict mass transfer coefficients more accurately than equations of Colazo *et al.* [equation (1)] and of Churchill [equation (2)]. The first represents the experimental data with a mean deviation of 13% and the second was shown to predict values which may differ by more than 15% from the experimental values.

An attempt to extend the theoretical model, recently formulated by Kobus and Wedekind [26], to describe combined forced and natural convection for vertical plates, proved unsuccessful because of the extreme

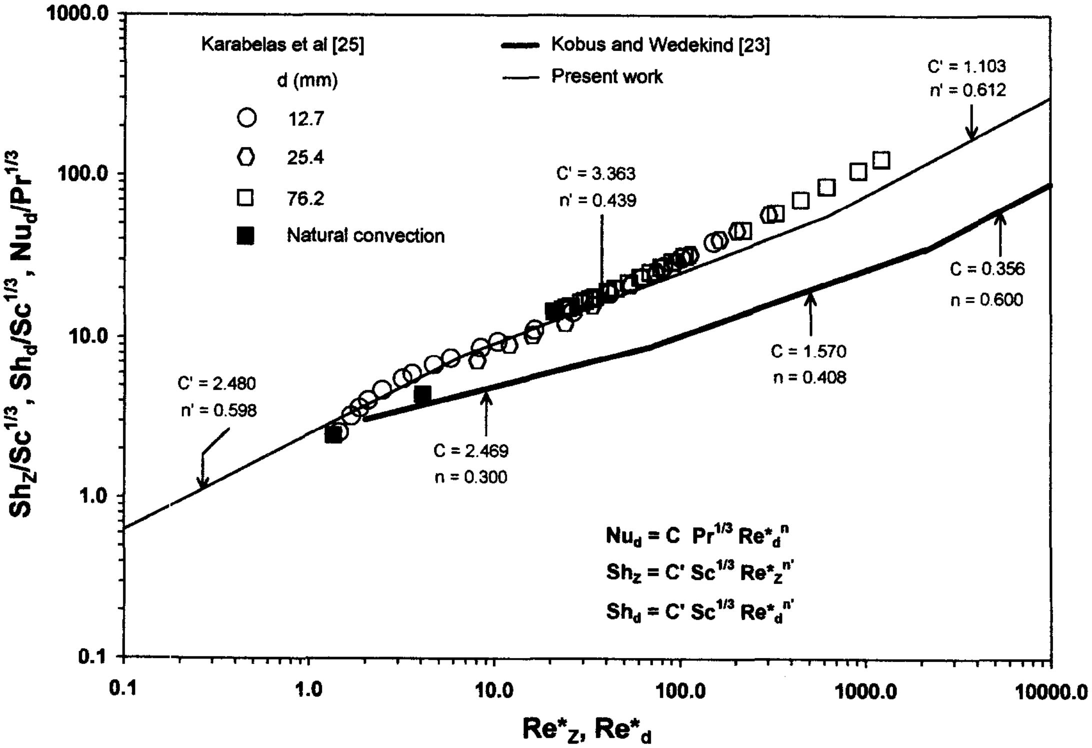


Fig. 12. Comparison of mass transfer correlations for structured packings with mass transfer in packed beds of spheres and heat transfer at vertical disks.

thinness of the diffusion layer in liquids of high Schmidt number. According to Kobus and Wedekind their model only would be applicable for $Pr < 10$.

## 9. CONCLUSIONS

Experimental investigation of mass transfer to regular packings carried out at low Reynolds numbers showed an appreciable influence of natural convection mass transfer, thus leading to an exhaustive study of this mode of convection.

Mass transfer correlations could be established for the different geometric configurations investigated. The equation

$$Sh_{L^*} = 0.247(Sc\,Gr_{L^*})^{0.3} \quad (20)$$

may be recommended as the most useful and general relationship for design purposes.

Besides, modeling the regular packing as an assemblage of inclined surfaces, an alternative method of predicting free convection mass transfer rates is presented.

Now, the availability of correlations describing pure natural and pure forced convection mass transfer in regular packings, allowed to apply the well known coupling rule to the mass transfer data corresponding to the low Reynolds number range.

Again, an alternative method of describing combined natural and forced convection mass transfer is presented. The use of a modified Reynolds number, which includes a term accounting for the buoyancy-induced velocity, yields the correlation

$$Sh_Z = 3.363 Sc^{1/3}\, Re_Z^{*\,0.439} \quad (19)$$

to be used in the low Reynolds number range.

The empirical mass transfer correlations developed for the pure free and pure forced convection modes can also be expressed with the aid of the modified Reynolds number.

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