

# Mass transfer variation within a packed bed and its relation to liquid distribution

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Received 1 July 2003; accepted 6 December 2003

## Abstract

The limiting current method is applied to characterize the local mass transfer in the axial, radial and angular directions throughout a packed bed of Pall rings. This is expressed in terms of the particle-to-fluid mass transfer coefficient of ferricyanide ions obtained from the limiting currents measured during its reduction to ferrocyanide at different locations in the bed. Particular focus is placed on the variability of the mass transfer coefficient with position in terms of the quantity  $MT_{VAR}$ , equivalent to the normalized deviation of the function  $Sh/Sc^{0.33}$ . The results confirm those reported by other researchers using more traditional liquid collection and tracer methods. Mass transfer coefficients throughout the bed closely follow normal distributions for the range of  $Re$  values investigated. When a multipoint distributor is used,  $MT_{VAR}$  is already low at the lowest flow rate ( $Re = 95$ ) considered and remains unchanged with any further increase in flow rate. In the case of a single-point distributor, on the other hand,  $MT_{VAR}$  decreases with increasing flow rate, but always remains significantly higher than that observed with the multipoint distributor. Moreover, the  $MT_{VAR}$  value depends on whether irrigated or full flow conditions prevail and on the cross-sectional area of the sampling electrodes, suggesting that local mass transfer variations are caused by local liquid velocity variations once the Pall rings are sufficiently wetted.

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**Keywords:** Local mass transfer variation; Limiting current method; Liquid distribution

## 1. Introduction

Analysis of liquid flow patterns is one of the main diagnostic tools used for evaluating the performance of packed beds. Although the final measure of performance is actually the mass transfer efficiency, the liquid flow conditions inside the column actually dictate how it performs. For this reason, observation and analysis of the local liquid flow rates are important for improving the design of packed columns. Typically, the measurement of liquid flow rate is straightforward and involves placing an array of liquid collectors at the outlet of the bed. This method has been used to great effect to yield quite detailed data [1], although more advanced techniques are now available [2]. However, despite its simplicity and apparent success, the method does have some shortcomings. Firstly, since the mass transfer coefficient is not measured directly, mass transfer rates can

only be inferred from liquid flow or local concentration measurements. Another main drawback is that liquid collection systems are limited to certain modes of packed bed operation due to their physical set-up. It is not possible to utilize this method for flow regimes other than irrigated liquid downflow, and even then the introduction of gas flow is not possible without major modifications.

In the current study, we report the use of the electrochemical limiting current method that both complements and extends the liquid collection studies. This approach overcomes some of the shortcomings of other techniques since it permits reaction rates and particle-to-fluid mass transfer coefficients to be directly determined (through measurement of current at sampling electrode packing pieces) and does so in a non-destructive manner without altering the packing surface shape and texture. Moreover, these values can be obtained instantaneously, thereby allowing transient effects or reactors operating under non-steady state conditions to be characterized. Unlike liquid collection techniques that permit simultaneous measurements in the radial direction but are restricted to sampling only at the bed outlet, there is essentially no limit to obtaining measurements simultaneously in the radial, axial and angular directions using the

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### Nomenclature

$a$	cathode area (m <sup>2</sup> )
$C_b$	bulk concentration of ferricyanide (mol m <sup>-3</sup> )
$d$	column diameter (m)
$d_e$	equivalent diameter of packing (m)
$D$	diffusion coefficient of ferricyanide in solution (m <sup>2</sup> s <sup>-1</sup> )
$F$	Faraday constant (96,487 C mol <sup>-1</sup> )
$i_L$	limiting current for ferricyanide reduction (A)
$k_i$	normalized concentration measured at $i$ th collector or sampling electrode
$k_L$	liquid-phase mass transfer coefficient of ferricyanide at solution/packing interface (m s <sup>-1</sup> )
$M_f$	maldistribution index based on volumetric flow rate
$M_k$	maldistribution index based on normalized concentration
$MT_{VAR}$	dimensionless function defined in Eq. (4)
$n$	total number of collectors or sampling electrodes in column
$q_i$	volumetric flow rate to $i$ th collector or sampling electrode in column (m <sup>3</sup> s <sup>-1</sup> )
$Re$	Reynolds number ( $Re = Ud_e/\nu$ )
$Sc$	Schmidt number ( $Sc = \nu/D$ )
$Sh$	Sherwood number ( $Sh = k_L d_e/D$ )
$U$	superficial velocity of liquid through column (m s <sup>-1</sup> )
$x/d$	dimensionless axial distance from liquid distributor
$z$	number of electrons transferred during reduction of each ferricyanide ion
$\nu$	kinematic viscosity of solution (m <sup>2</sup> s <sup>-1</sup> )
$\langle \rangle$	mean value of function

electrochemical method. This allows mass transfer to be characterized on both macroscopic and microscopic scales. Finally, the electrochemical method can characterize mass transfer and liquid distribution during upward flow and both full and irrigated downward flow modes of operation.

The study of the flow distribution of liquids in packed beds has received much attention. Several groups have obtained excellent data. Hoek and co-workers [1,3,4] measured liquid distributions at many locations within the outlet cross-section of packed beds for a wide variety of bed heights, packing materials, packing sizes and liquid distributors. A similar study was performed by Al-Samadi et al. [5] who also varied the number of nozzles on the liquid distributor. They observed a decrease in the maldistribution index:

$$M_f = \frac{1}{n} \sqrt{\sum_{i=1}^n \left( \frac{q_i - \langle q \rangle}{\langle q \rangle} \right)^2} \quad (1)$$

with an increasing number of nozzles as long as fewer than 12 m<sup>-2</sup> were installed. However, beyond this number of nozzles, no further improvement in  $M_f$  was obtained although it still did not reach zero, or perfect distribution. A maldistribution index of about 0.25 still existed in the bed between locations.

More recently, Kouri and Sohlo [6] measured the liquid distribution in a packed bed, focusing on the development of wall flow. They also attempted to measure the mass transfer behaviour using thermocouples to measure temperature differences due to water evaporation. Due to the changing relative humidity and temperatures of both phases, this method required a great deal of interpretation to extract the mass transfer coefficients and accordingly only the raw temperature data were presented. They found that the variation in temperatures in the column varied directly with  $M_f$ . The method of measuring temperature profiles was adapted from the work of Stichlmair and Stemmer [7], who used several hundred thermocouples to measure the temperature profiles caused by water evaporation in a packed bed. They also did not extract mass transfer coefficients from the data, showing instead the isotherms in the column. Both studies provided very clear qualitative pictures of the mass transfer behaviour under different liquid distribution conditions. However, they still suffer from not directly measuring mass transfer rates.

Groenhof [8,9] measured the liquid distribution in conjunction with stripping acetone from water to determine the degree of liquid maldistribution in a packing. It was found that  $M_f$  and the maldistribution index  $M_k$  based on normalized concentration:

$$M_k = \left( \sum_{i=1}^n \left( \frac{(k_i - \langle k \rangle)^2}{n} \right) \right)^{0.5} \quad (2)$$

were 0.39 and 0.17, respectively. Although this work provided an excellent comparison between liquid distribution and concentration variations, its main focus was on the effect of column diameter on the scaling-up of packed beds. It did not involve a systematic analysis of the local behaviour (i.e. changes with packing height) nor did it study the effect of different inlet liquid distribution modes.

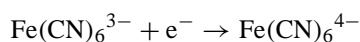
Recently, Gostick et al. [10] used an electrochemical method to directly measure the axial, radial and angular dependence of local particle-to-fluid mass transfer coefficients within a packed bed of Pall rings for both irrigated liquid downflow and full liquid upflow operation. The axial profiles were based on measurements at nine electrodes placed at different radial and angular locations at each of six positions along the length of the column. These experiments revealed that the mass transfer rate showed a maximum at an axial distance of 1.5 column diameters when the liquid was fed into the column using a multipoint distributor and at a distance of 2.5 column diameters when a single-point distributor was used. In both cases, the average mass transfer coefficient at each axial position eventually reached a constant value further down the column.

In this study, we once again report on the use of the electrochemical method, but now focus on the variability of mass transfer coefficients depending on location within a packed column. The variation in values obtained over 54 different locations throughout the column is determined at various liquid flow rates using both single and multipoint liquid distributors. The data are also analyzed to determine the axial and radial dependence of this variability. Experiments will be carried out to gain some insight into the origin of this variability. To our knowledge, this is the first study to report on the variation of mass transfer coefficients as opposed to liquid flow rates or local concentrations. Such information is potentially important since it can provide further insight into the maldistribution of liquid flow through packed columns. Typically, mass transfer in packed beds has been characterized on the basis of measurements made at only a few locations within columns. The variability of mass transfer coefficients with bed position determined in this study will also enable us to assess how representative of the true overall performance are measurements at only a few locations and the most suitable locations for such measurements.

Comparisons will be made between the mass transfer coefficient distributions obtained in this study to liquid flow distributions obtained by other researchers using the liquid collection approach. This will provide information on the relationship between the liquid flow rate and mass transfer coefficient distributions within the bed. The limiting current method also allows all modes of operation of a given packed column to be investigated. Consequently, the distribution of mass transfer coefficients under full upward flow conditions will be measured and serve as a baseline to which the distributions obtained during irrigated downward flow operation can be compared.

## 2. Experimental

Particle-to-fluid mass transfer coefficients were obtained at various locations in a packed bed using the electrochemical technique. General aspects of the method are given by Selman and Tobias [11]. The electrochemical reaction system chosen for our study was the ferricyanide/ferrocyanide redox couple:



This choice has several advantages: this system is well understood, exhibits fast electrode kinetics and does not involve any solid reactants or products that would alter the packing surface. The solution continuously passed through the packed bed contained  $3.8 \text{ mol m}^{-3} \text{ Fe}(\text{CN})_6^{3-}$ ,  $4.0 \text{ mol m}^{-3} \text{ Fe}(\text{CN})_6^{4-}$  and  $500 \text{ mol m}^{-3} \text{ NaOH}$ . The column was configured so that the bed contained cathodic and anodic layered sections. At specific locations within each section were placed a small number of nickel-coated Pall rings electrically connected to a power supply. Each of these electrodes was surrounded by insulating polymer-coated Pall rings to

prevent short-circuiting. Each electrode layer in turn was separated from the adjacent one with another layer of insulating Pall rings. To characterize mass transfer within the bed, voltage was applied to the circuit and the resulting current was measured. Sufficient voltage was applied to ensure that ferricyanide reduction at the cathodes was mass-transfer limiting. In this way, the measured limited current  $i_L$  was proportional to the mass transfer coefficient for ferricyanide at the solution/packing interface via the expression:

$$k_L = \frac{i_L}{azFC_b} \quad (3)$$

where  $a$  is the cathode area,  $z$  the number of electrons transferred by the electrode reaction for each ferricyanide ion reduced,  $F$  the Faraday constant and  $C_b$  the bulk concentration of ferricyanide in the solution flowing through the packed bed. A more detailed discussion of the experimental set-up and procedures is given elsewhere [10,12].

The apparatus consisted of a packed bed of 25 mm Pall rings with a column diameter of 0.30 m. A packed bed height equal to 5.5 column diameters was tested. Two types of liquid distributors were used: an 'X'-type distributor with 16 nozzles, giving a nozzle density of  $220 \text{ m}^{-2}$  and a single-point distributor placed at the radial centre of the bed. Liquid flow rates between 9350 and  $56,120 \text{ kg m}^{-2} \text{ h}^{-1}$  were used, providing a range of Reynolds numbers between 95 and 560. The Schmidt number was held constant at approximately 1500. Fig. 1 shows the schematic diagram of the apparatus. The column could be run in full liquid upflow or irrigated downflow mode. Although a gaseous component was not involved in this study, the set-up could

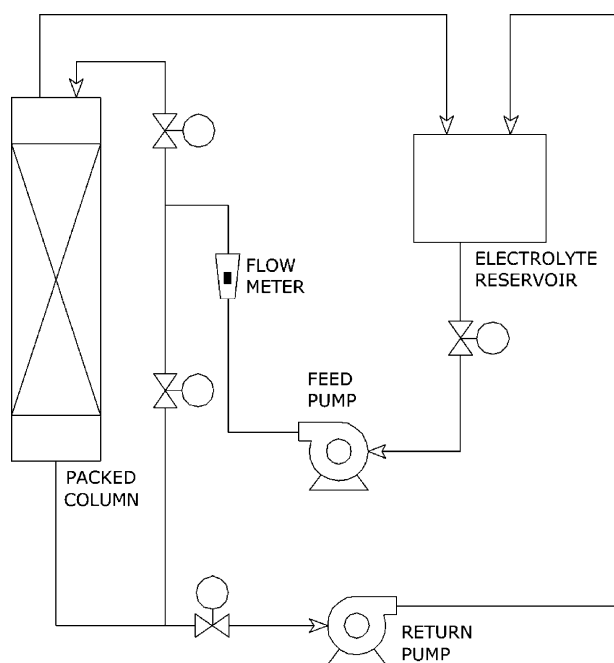


Fig. 1. Schematic diagram of experimental set-up to measure local particle-to-fluid mass transfer coefficients in packed beds.

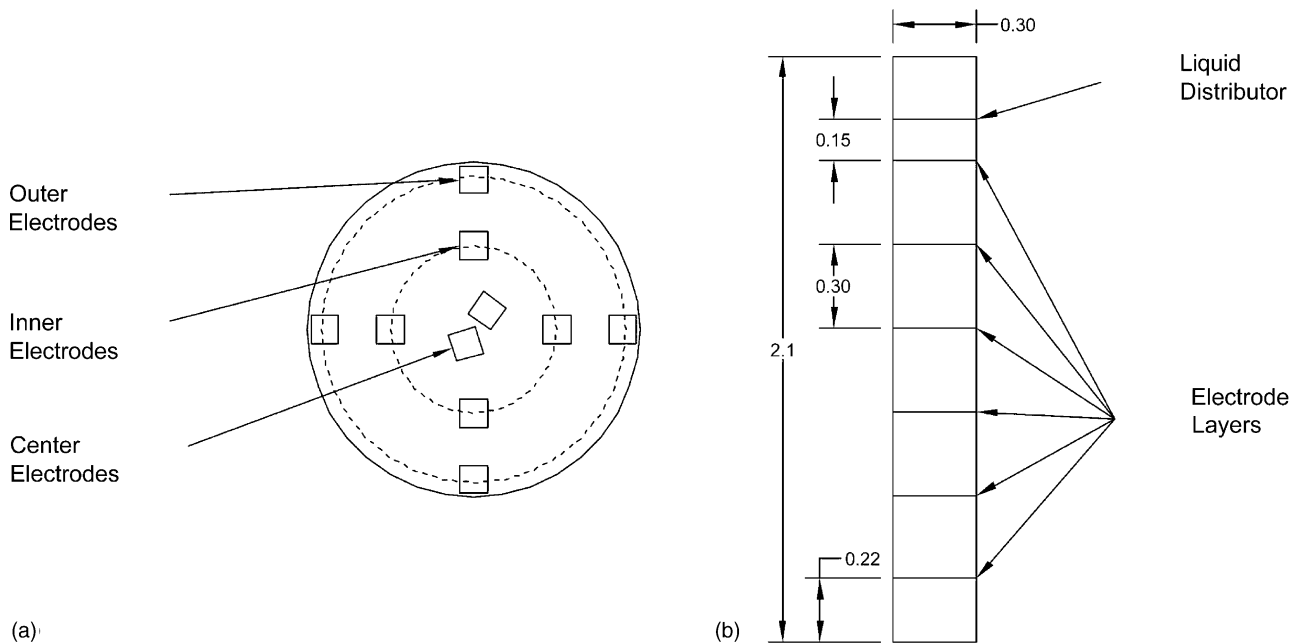


Fig. 2. Schematic diagram showing the radial (a) and axial (b) placement of nickel-plated Pall rings that serve as electrodes to measure limiting currents. The dimensions are given in metres.

be easily modified to allow a gas to be fed into and/or collected from the bed.

Particle-to-fluid mass transfer coefficients were obtained at 54 locations in the bed over six axial positions. At each axial location, nine electrodes were arranged as shown in Fig. 2a. Two Pall rings electrically connected to each other served as an electrode in the centre of the column at each axial position to provide a larger sampling surface area. This was found to yield a more representative measure of the average flow through the central portion since the value could not be averaged as for the inner and outer ring sections. Obviously, in calculating  $k_L$  from Eq. (3), the area used for the central electrode was twice that for the other electrodes. More concerning the effect of sampling area on the measurements is discussed in Section 3.1. The six axial positions were 30 cm apart, starting at 15 cm below the liquid distribution point at the axial position  $x/d = 0.5$  (Fig. 2b).

An important feature of the set-up allows the multipoint liquid distributor to be rotated in the angular direction. Some of the data presented in this study represents a combination of measurements obtained for different orientations of the distributor with respect to the array of electrodes. In addition, analysis of the data for each angular position enables the effect of distributor position relative to that of the electrodes to be determined. This effect was found to be particularly important for the electrodes near the top of the column.

### 3. Results and discussion

The data obtained at each electrode can be combined in several ways to reveal different trends and behaviour. For in-

stance, data from each electrode in the column can be combined to give a measure of the overall behaviour of a column. Alternatively, data from electrodes at specific locations can be presented to reveal local phenomena. The results of these analyses are presented in Sections 3.1 and 3.2.

#### 3.1. Distribution of mass transfer coefficients over the entire bed

It is of interest to investigate the distribution of mass transfer coefficient values measured in a packed bed. The utility of overall mass transfer correlations depends on the statistical distribution of mass transfer coefficients throughout the bed being distributed normally with a small variance. A small variance indicates that relatively homogeneous liquid flow is occurring throughout the column. Furthermore, the overall statistical distribution in the bed should be representative of local conditions in the column. It has been found that these criteria are only partially met in the column used in the present experiment.

Many researchers have pointed out that the random nature of packed beds should result in a normal distribution for mass transfer [13,14] and heat transfer [15,16] coefficients. Furthermore, Sederman et al. [2] showed that the liquid velocities through the pores of a packed bed also vary in a statistically normal way. A histogram of the frequency distribution of mass transfer coefficients obtained in our column is compared to a normal distribution based on the mean and variance of the experimental data in Fig. 3. The good agreement indicates that the measured values follow a normal distribution. Similar fits were obtained for other flow rates investigated.

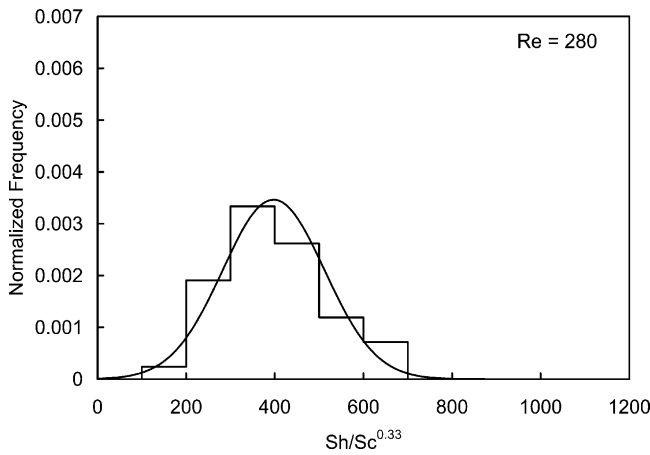


Fig. 3. Histogram of the frequency distribution of local particle-to-fluid mass transfer coefficients over the entire packed bed as a function of  $Sh/Sc^{0.33}$  for downward irrigated flow at  $Re = 280$  using the multipoint liquid distributor. The solid curve represents the normal frequency distribution predicted on the basis of the mean and variance of the experimental data.

Normal distribution curves obtained for the various flow rates investigated are superimposed in Fig. 4. Each curve is based on the mean and variance experimentally observed at each flow rate. It can be seen that the variance increases with flow rate. However, this trend can be misleading because the variance is not normalized to account for the increasing mean (i.e. larger numbers have larger variances). A function that accounts for this effect by normalizing the deviation can be defined as follows:

$$MT_{VAR} = \frac{1}{n} \sum_{i=1}^n \left( \frac{|(Sh/Sc^{0.33})_i - \langle Sh/Sc^{0.33} \rangle|}{\langle Sh/Sc^{0.33} \rangle} \right) \quad (4)$$

Obviously, the lower the value of  $MT_{VAR}$  is, the more uniform is mass transfer throughout the bed. In Fig. 5, this function is plotted versus  $Re$  for the cases of downward

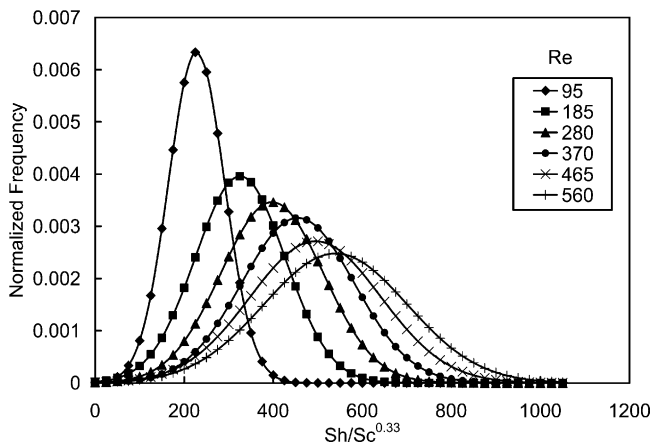


Fig. 4. Normal frequency distributions for local particle-to-fluid mass transfer coefficients over the entire packed bed as a function of  $Sh/Sc^{0.33}$  for downward irrigated flow at various  $Re$  values using the multipoint liquid distributor. Each curve is based on the mean and variance of the experimental data at the corresponding  $Re$ .

flow using the single-point and multipoint distributors and full liquid upward flow. It remains essentially constant in the case of the multipoint distributor and full upward liquid flow, whereas it decreases with increasing flow rate when the single-point distributor is used.

It is interesting that  $MT_{VAR}$  does not decrease with increasing  $Re$  for the multipoint liquid distributor since a higher liquid loading would be expected to more completely spread liquid throughout the bed and wet the packing, leading to more homogeneous flow and conditions approaching that of full liquid flow. According to the data, this explanation is only partially correct. An increase in liquid homogeneity would account for the improvement with increasing  $Re$  for the single-point liquid distributor since more liquid is available to spread away from the point of distribution. It also explains why the multipoint liquid distributor displays much lower  $MT_{VAR}$  values than does the single-point distributor since the liquid is discharged more equally over the bed and, in effect, pre-spreads the liquid. However, it does not explain why the multipoint liquid distributor shows no decline in  $MT_{VAR}$  with increasing liquid load. This may be due to the fact that the particles are already completely wetted and so any additional liquid fed to the column will have no further effect. Similar findings were reported by Al-Samadi et al. [5] for liquid distribution with a varying number of nozzles on the liquid distributor. The data in Fig. 5 show that the  $MT_{VAR}$  values for the single-point liquid distributor are decreasing toward the limiting value obtained with the multipoint distributor as  $Re$  rises. This effect is elucidated further in Section 3.2 where the effect of axial distance from the liquid distributor is considered.

However, it is likely that liquid spreading and wetting alone do not control the variation of mass transfer throughout the bed. As shown in Fig. 5, the full upward liquid flow regime, which of course has complete wetting, yields a lower value of  $MT_{VAR}$  than the apparent limiting value obtained with the multipoint liquid distributor. Since the

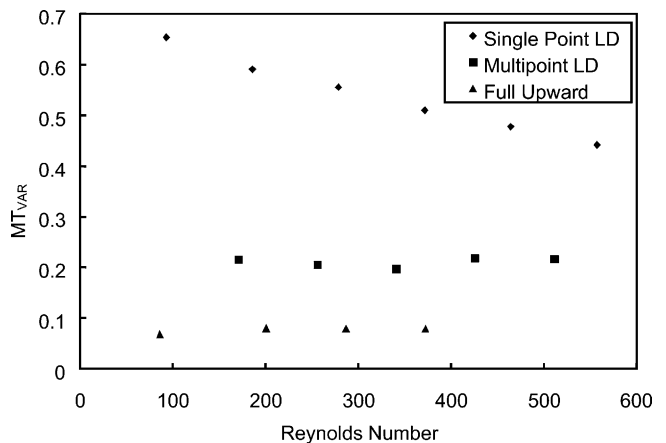


Fig. 5. Effect of  $Re$  on  $MT_{VAR}$  over the entire packed bed for cases of a single-point liquid distributor, multipoint liquid distributor and full upward liquid flow.



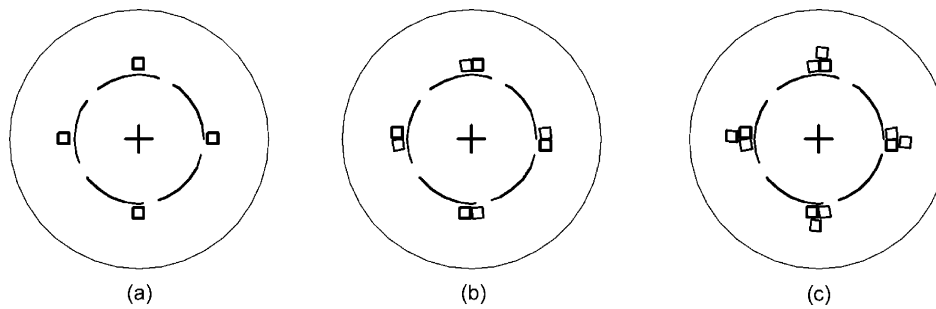


Fig. 6. Placement of electrodes for experiments to assess the effect of sampling area on the variance of measured  $MT_{VAR}$  values.

mass transfer coefficient is a function of both wetted area and liquid velocity, it is probable that variations in the liquid velocity are also contributing to the variations in mass transfer coefficient. Considering the flow patterns present in each regime this explanation seems most likely. Full upward liquid flow yields regions of higher velocity in the constrictions between packing pieces while irrigated flow is characterized by high velocity streams of liquid flowing through the bed, i.e. channelling. The non-uniformity of liquid velocity due to the constrictions in full liquid flow would be small compared to the difference in velocity between a channel and a stagnant zone in irrigated flow. This would explain the differences in  $MT_{VAR}$  for the two regimes.

A test of this explanation was conducted by determining the variability of the mass transfer coefficient measurements over the four angular positions as a function of the number of electrode packing pieces sampled at each location. The objective was to determine if the  $MT_{VAR}$  would be reduced by providing a larger electrode sampling area and a more representative overall flow pattern, thereby reducing the effects of localized stagnant zones and liquid channels. The different arrangements examined are shown in Fig. 6. For the configurations shown in Fig. 6b and c, each electrode piece was in electrical contact with the other electrode(s) at the same location.

Fig. 7 shows the variation of  $MT_{VAR}$  over the four angular positions in the inner ring section at  $x/d = 1.0$  as a function of the number of electrode pieces at each position when the multipoint liquid distributor is used. A significant decrease in  $MT_{VAR}$  occurs when the number of packing pieces is increased. Notice that as the number of packing pieces increases, the value of  $MT_{VAR}$  tends to level and approaches 0.07, which is identical to the value for full liquid flow shown in Fig. 5. Similar behaviour was reported by Porter et al. [17] upon increasing the sample collection area in their measurement of liquid volume spread in a randomly packed bed.

This is an important result since it indicates that the major cause of mass transfer deviations in an irrigated packed bed are velocity variations and not particle wetting, given that the limiting homogeneity has been reached. Although it is possible to reduce the value of  $MT_{VAR}$  in a bed either through improved liquid distributor designs, better perform-

ing packing elements or other means, it is not possible to completely eliminate variations in mass transfer coefficient values. Velocity fluctuations caused by the segregated nature of flow through a random packing will always occur.

### 3.2. Axial and radial variation of mass transfer coefficients

The results in Section 3.1 were based on combining data from every electrode throughout the bed. It will also be useful to examine the original data obtained from each electrode and analyze the effect of position in the bed on the variability of the mass transfer coefficient. Care must be taken when analyzing the results on a local scale. Positional dependence, particularly in the axial direction, has a significant effect on the liquid distribution due to liquid build-up on column walls [18–20]. Mass transfer coefficients have also been shown to vary predictably with position [10,20]. As a result, not all variations are due to random processes. It should be noted that the value of  $\langle Sh/Sc^{0.33} \rangle$  used to calculate  $MT_{VAR}$  at local positions according to Eq. (4) is the mean value based on measurements throughout the entire bed.

Fig. 8 shows the change in  $MT_{VAR}$  as a function of axial position in the bed with the multipoint liquid distributor

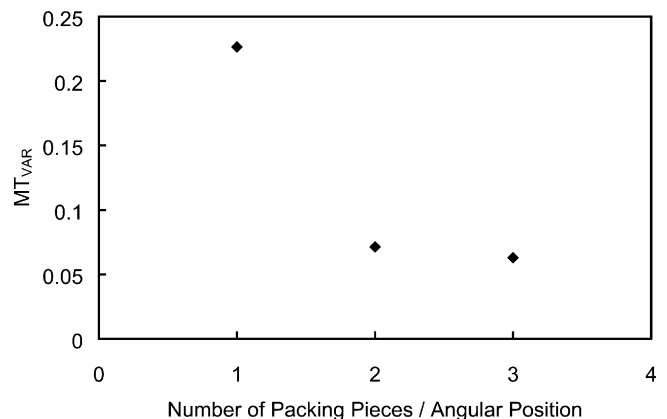


Fig. 7. The variation of  $MT_{VAR}$  over the four angular positions in the inner ring section at  $x/d = 1.0$  for irrigated downward flow as a function of the number of electrode pieces at each position when the multipoint liquid distributor is used.

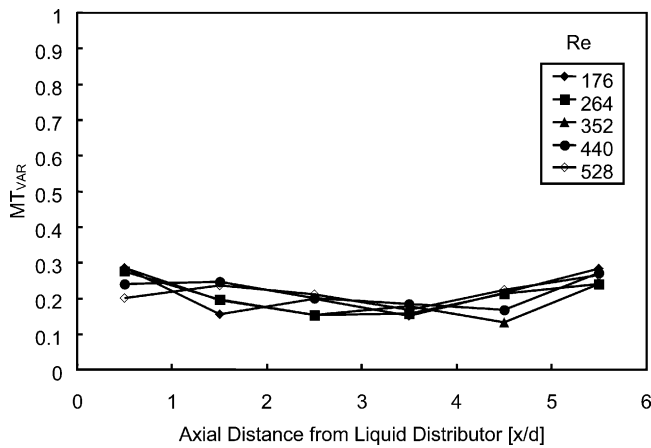


Fig. 8. Effect of axial position on  $MT_{VAR}$  for downward irrigated flow at various  $Re$  values using the multipoint liquid distributor. Each  $MT_{VAR}$  value is based on the measurements at the nine electrodes in the radial and angular locations shown in Fig. 2a for each axial position.

over a range of  $Re$  values. Each data point shown is based on the separate measurements at the nine electrodes in the radial and angular locations shown in Fig. 2a for each axial position. Due to the good liquid feed distribution resulting from the use of the multipoint distributor, the value of  $MT_{VAR}$  does not change as the liquid flows down the column. Furthermore, volumetric flow rate has very little effect on the variability of mass transfer coefficients. Hoek et al. [3,4] also observed that the maldistribution index  $M_f$  did not vary with axial position for a variety of packing types and inlet flow rates. It should be noted that they investigated the axial dependence by carrying out separate experiments in which liquid was collected at the outlet of beds with varying heights. This should be contrasted with the current study where the axial dependence was determined through

simultaneous measurements over one column during a single experimental run.

The data from numerous runs with different relative angles between the liquid distributor arms and the nine sampling electrodes have been combined to produce the results shown in Fig. 8. This produces an averaged picture of the flow through a bed with a good inlet distribution. Examination of the data according to angular orientation of the liquid distributor with respect to the electrodes leads to a different picture. Fig. 9 shows the effect of axial position on  $MT_{VAR}$  obtained for two liquid distributor positions. The results obtained when the liquid is dispensed directly over the sampling electrodes are presented in Fig. 9a, whereas the results obtained when they are  $45^\circ$  apart are shown in Fig. 9b. Comparison of the two sets of data reveals that even when a multipoint liquid distributor is used, the  $MT_{VAR}$  is not necessarily a minimum just below the distribution point as indicated by the averaged values in Fig. 8. When the liquid is dispensed directly over the sampling electrodes, one gains the impression that the bed is fully wetted since the corresponding  $MT_{VAR}$  values remain constant. However, the results in Fig. 9b indicate that poorly irrigated areas exist directly below the distributor and some angular dispersion is required before the electrodes become completely wetted. This effect corrects itself quite quickly and  $MT_{VAR}$  reaches the minimum value after about 1.5 column diameters.

An interesting phenomenon that can be seen from Fig. 9a is that the  $MT_{VAR}$  does not change as liquid flows down the column. An increase in  $MT_{VAR}$  might be expected since there is an overall loss of liquid from the area of the electrodes as it migrates to the drier areas of the bed. In fact, the  $MT_{VAR}$  remains constant probably due to the existence of a limiting homogeneity discussed earlier. There is a surplus of liquid in an already wetted area beyond that required to

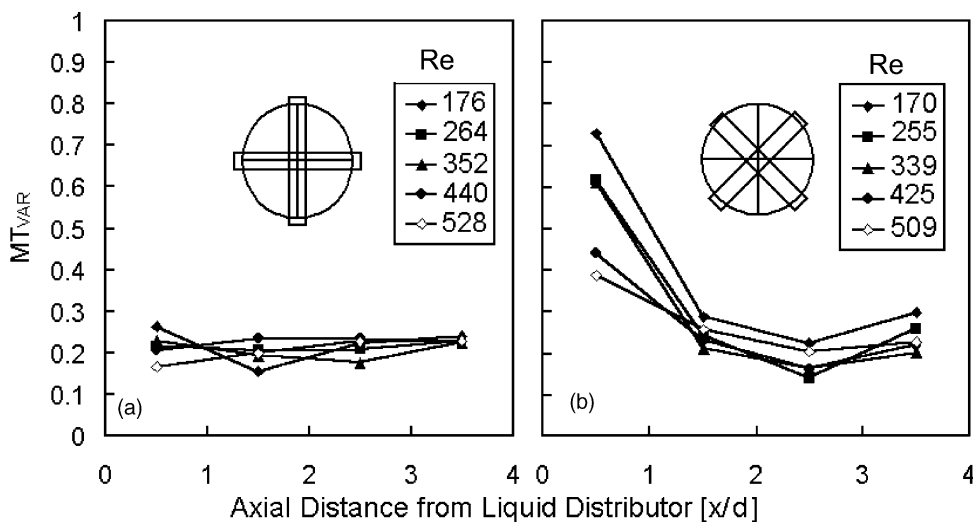


Fig. 9. Effect of axial position on  $MT_{VAR}$  for downward irrigated flow at various  $Re$  values when the multipoint liquid distributor is oriented  $0^\circ$  (a) and  $45^\circ$  (b) from the electrode positions. Each  $MT_{VAR}$  value is based on the measurements at the nine electrodes in the radial and angular locations shown in Fig. 2a for each axial position.

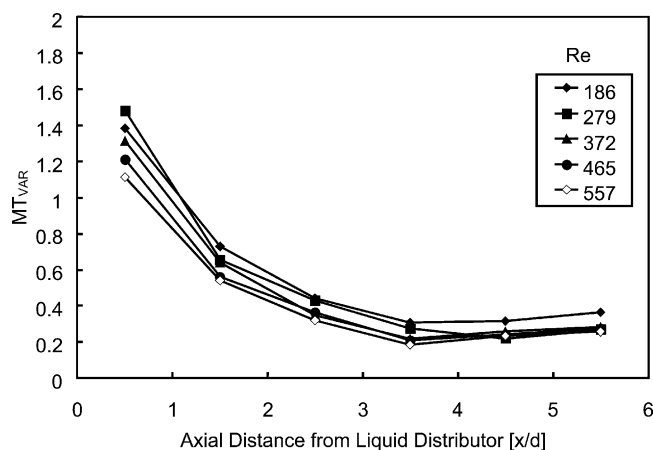


Fig. 10. Effect of axial position on  $MT_{VAR}$  for downward irrigated flow at various  $Re$  values using the single-point liquid distributor. Each  $MT_{VAR}$  value is based on the measurements at the nine electrodes in the radial and angular locations shown in Fig. 2a for each axial position.

establish a limiting homogeneity and so liquid lost from an area has no effect on  $MT_{VAR}$ .

The axial dependence of  $MT_{VAR}$  obtained with the single-point liquid distributor shows much different behaviour, as evident in Fig. 10. Very high values of  $MT_{VAR}$  are obtained at the top of the column. Just below the single-point liquid distributor, the centre packing is flushed with liquid, while the outer section is practically dry. As the liquid flows down the column and spreads out over the packing, the outer section receives more liquid so that the flow there comes into line with that through the inner section, thereby reducing the overall variation in the bed. This process is related to the development of equilibrium wall flow discussed previously by many workers [6,21,22].

Note that at the bottom of the column the value of  $MT_{VAR}$  matches that obtained with the multipoint liquid distributor. This is more evidence in support of the existence of the limiting homogeneity discussed in Section 3.1. Similar behaviour was noticed by Porter and Jones [23] using gas flow with a tangential inlet to give a poor feed distribution. Gunn and Al-Saffar [24] went one step farther and introduced disproportionate amounts of liquid at the outer radius of the bed. Their results show a similar ability of the packing to redistribute liquid.

The local analysis provides a useful breakdown of the column behaviour. From the local data, it is possible to view the effectiveness of a liquid distributor design, measure the ability of a packing material to redistribute liquid or analyze the effect of other geometric factors such as packing size or column diameter.

In view of the results in Fig. 7, our use of the set-up shown in Fig. 2 potentially influences the values of  $MT_{VAR}$  obtained since two Pall rings comprise the central electrode and only one for each of the remaining electrodes. As discussed in Section 2, such a set-up was implemented to improve the reproducibility of the current measurements at the

central electrode. Consequently, the set-up represents a compromise between improved reproducibility and a potentially lower  $MT_{VAR}$  value. To assess the effect of the set-up on  $MT_{VAR}$ , the data on which Figs. 8–10 are based have been re-analyzed excluding the measurements at the central positions. This resulted in only very small changes in the  $MT_{VAR}$  values and no qualitative changes whatsoever in the trends shown in Figs. 8–10. This should not be surprising since the measurement at each central electrode is combined with the measurements at the eight other radial locations at each axial position to yield each of the data points in these figures.

#### 4. Conclusions

An electrochemical technique in which the limiting current for a redox couple is measured has been successfully employed to measure particle-to-fluid mass transfer coefficients simultaneously at a large number of locations within a packed bed. A number of findings obtained confirm those reported previously by other researchers using more traditional liquid collection and tracer methods. It has been shown that the average variation in the mass transfer coefficient decreases to a minimum limiting value when either a multipoint liquid distributor provides a good feed liquid distribution or a single-point liquid distributor is used and the liquid spreads over the packing as it flows down the column. Additionally, it has been found that increasing liquid flow rate does not decrease the average variation in the mass transfer coefficient when a multipoint distributor is used, but does so when a single-point distributor is used. This suggests that additional liquid load to the column helps the poorly distributed liquid spread out and reduce the value of  $MT_{VAR}$ . Together, these trends suggest that the packing reaches a nearly fully wetted state. However, it was observed that a  $MT_{VAR}$  value of about 0.2 in the case of irrigated downward flow using a multipoint distributor still existed in the bed compared to the case of full upward liquid flow conditions where a  $MT_{VAR}$  of only 0.07 is measured. This difference was attributed to the more extreme nature of velocity variations in an irrigated bed, caused by channelling through the packing. A value of 0.07 was obtained for irrigated flow by increasing the cross-sectional area of the sampling electrodes and thereby contacting a more averaged portion of the flow. This provides evidence that the variability of mass transfer coefficient with location are caused by local liquid velocity differences once a limiting wetting state has been reached.

A main advantage of the limiting current method is that it enables reaction rates and mass transfer coefficients to be instantaneously and simultaneously measured at an essentially unlimited number of locations in the axial, radial and angular directions. It can also be used in a wider range of flow regimes, including full liquid and gas-continuous flows, than other methods. Since a main cause of mass transfer coefficient variability appears to be variations in local liquid velocity, it has been possible to infer the effect



of liquid flow distribution on mass transfer throughout the bed. A next logical step would be to combine the electrochemical technique with traditional liquid collection methods to directly link liquid flow patterns and mass transfer. An effort in this direction was made by Groenhof [8,9], although local concentrations and not mass transfer coefficients were measured within a packed bed. Thus, such an approach cannot directly measure the link between liquid flow and mass transfer since assumptions regarding the liquid flow patterns would still be required to relate local concentrations to mass transfer. On the other hand, the combination of the electrochemical method utilized in the present study and liquid collection would allow the relationship between liquid flow and the mass transfer coefficient to be directly measured without the need for any such assumptions.

### Acknowledgements

Financial support from the Natural Sciences and Engineering Research Council of Canada (NSERC) for this project is greatly appreciated. Gratitude is also expressed to Koch-Glitsch, Uxbridge, Ont., Canada for donating Pall rings.

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