

## Mass transfer between a liquid and an array of discs in a cylindrical container. Part II: Combined pumped flow and rotation

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#### Abstract

The work is a continuation of an electrochemical mass transfer study which employed a cell with alternatingly spaced annular and full discs. Pumped liquid flow through the cell is now combined with rotation of the full discs. The influence of the different parameters on mass transfer at four characteristic surfaces in a cell element is analysed, and an empirical correlation is obtained for each surface. In the entrance region, where the radial flow is divergent, mass transfer control is mixed, while it is controlled by rotation in the convergent exit channel.

#### List of symbols

- $A_{\rm e}$  electrode surface area (m<sup>2</sup>)
- $C_{\infty}$  concentration of ferricyanide ions (mol m<sup>-3</sup>)
- *D* molecular diffusion coefficient (m<sup>2</sup> s<sup>-1</sup>)
- *e* hydraulic radius (m)
- *F* Faraday constant (96 500 C mol<sup>-1</sup>)
- *h* half-distance between two discs (m)
- *H* distance between two discs, 2h (m)
- $I_{\rm L}$  limiting diffusion current (A)
- $\bar{k}_{d}$  mean mass transfer coefficient (m s<sup>-1</sup>)
- *N* rotation velocity (rpm)
- $Q_V$  volumetric flow rate (m<sup>3</sup> s<sup>-1</sup>)
- $R_0$  radius of the full disc (m)
- $R_1$  inner radius of the annular disc (m)
- $R_2$  inner radius of the cell (outer radius of the annular disc) (m)

#### 1. Introduction

In Part I [1] mass transfer between a liquid and discs arranged in a cylindrical cell (Figure 1) was studied in the two limiting situations of pumped flow through a stationary arrangement, and rotation of the full discs. The electrochemically measured average mass transfer coefficients between the liquid and the four surfaces 1 to 4 of a modulus (Figure 1) were analysed and empirically correlated.

It is known that the space-time yield of electrochemical reactors is improved when high mass transfer

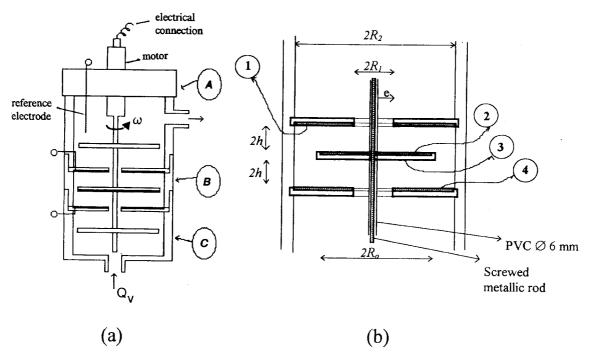
- $Re_{C}$  channel Reynolds number used by Jansson,  $(Q_{v}/Hv)$
- $Re_{\rm H}$  rotational Reynolds number based on H,  $(\omega H^2/\nu)$
- $Re_{\rm m}$  channel Reynolds number used by Kreith,  $(Q_{\rm v}/(2\pi Hv))$
- $Re_{\rm R}$  rotational Reynolds number based on R,  $(\omega R^2/v)$
- *Ro* Rossby number,  $(Re_C/Re_R)$
- Sc Schmidt number, (v/D)
- $\overline{Sh}$  mean Sherwood number,  $(2h\bar{k}_{\rm d}/D)$
- Ta Taylor number,  $(h^2\omega/v)^{0.5}$

Greek symbols

- $\beta$  exponent
- $\mu$  dynamic viscosity (kg m<sup>-1</sup> s<sup>-1</sup>)
- *v* kinematic viscosity (m<sup>2</sup> s<sup>-1</sup>)
- $\rho$  density (kg m<sup>-3</sup>)
- $\omega$  angular velocity (s<sup>-1</sup>)

coefficients at the working electrode are attained while volumetric flow rates are small [2]. This signifies that the mass transfer coefficient has to be increased by other means than the flow rate; one of the possible means is electrode rotation. This was the basic idea in the Couette–Taylor–Poiseuille electrochemical reactor [2– 4], and of the 'Eco-cell' proposed for metal recovery from dilute solutions [5]. Another electrochemical reactor which combines the rotation of one electrode and forced flow, in order to obtain high space-time yields, that is, high values of the conversion per pass through a reactor having a small volume, is shown in Figure 1. In





*Fig. 1.* Experimental cell: (a) general view; (b) view of the four surfaces in the modulus. Geometrical parameters:  $2R_0 = 50$ , 54 and 56 mm;  $2R_1 = 12$ , 16 and 20 mm;  $2R_2 = 60$  mm; 2h = 2, 4, 6 and 8 mm; e = 3, 5 and 7 mm.

the case of the 'pump cell', which only contains a rotating disc and a stationary disc, the flow rate is induced by rotation [6].

The present work concerns mass transfer to surfaces 1 to 4 of the cell in Figure 1 except that pumped flow is now combined with rotation of the full discs (surfaces 2–3). Such an arrangement has been previously studied as a rotating-disc contactor for liquid–liquid extraction [7]. Under turbulent conditions, intense recirculation between the rotating and the stationary discs provides thorough mixing of the two phases and, creates consequently, an efficient extraction process.

#### 2. Hydrodynamics and mass transfer from the literature

The system now considered has two limiting cases studied previously by the present authors: rotation of a disc in a closed cylinder [8] and pumped liquid flow alone between stationary discs [1].

The flow structure resulting from the combination of a pumped flow with the rotation of a disc opposite to a stationary disc was studied by several authors, especially in the case of single phase fluids. Recirculation is generated in the interdisc space, as a result of the fact that the rotating disc puts energy into the fluid by moving it towards the periphery, while energy is dissipated on the stationary disc [7]. The modification suffered by the flow structure depends on the radial flow direction between the discs (inflow or outflow). The works of Adams and Szeri [9], Szeri [10], Prakash [11], Thomas [12] and Peres [13] were devoted to such a complex flow, the study of which lies beyond the scope of our work. To estimate which mode exerts predominant influence, the channel Reynolds number,  $Re_C$ , and the rotation Reynolds number,  $Re_R$ , are combined in the Rossby number  $R_0 = Re_C/Re_R$  [10].

Average mass transfer rates between a fluid and the discs, in the presence of forced flow were studied by the groups of Kreith and Jansson. Based on measurements using the naphthalene dissolution method, Kreith [14] proposed an empirical correlation for mass transfer between a flowing gas and a rotating disc: the Sherwood number was expressed as function of the rotation Reynolds number, Re<sub>R</sub>, and the channel Reynolds number,  $Re_{\rm m} = Re_{\rm C}/(2\pi)$ . The research team of Jansson determined electrochemically mass transfer coefficients in view of the development of the pump cell. Jansson and Ashworth [15] proposed empirical correlations of the type  $\overline{Sh} = f(Re_{\rm C}; Re_{\rm R};$  geometrical parameters) for the rotating and the fixed disc. They observed that channel flow is dominant when  $Re_{\rm C}/Re_{\rm R} > 1$ ; they also measured distributions of local mass transfer coefficients over the discs, showing an entry effect. Their study was continued by Jansson and Marshall [16], and also by Groroghchian et al. [17] who considered a convergent flow (or inflow) and proposed empirical correlations giving the average Sherwood number, together with experimental distributions of local mass transfer coefficients.

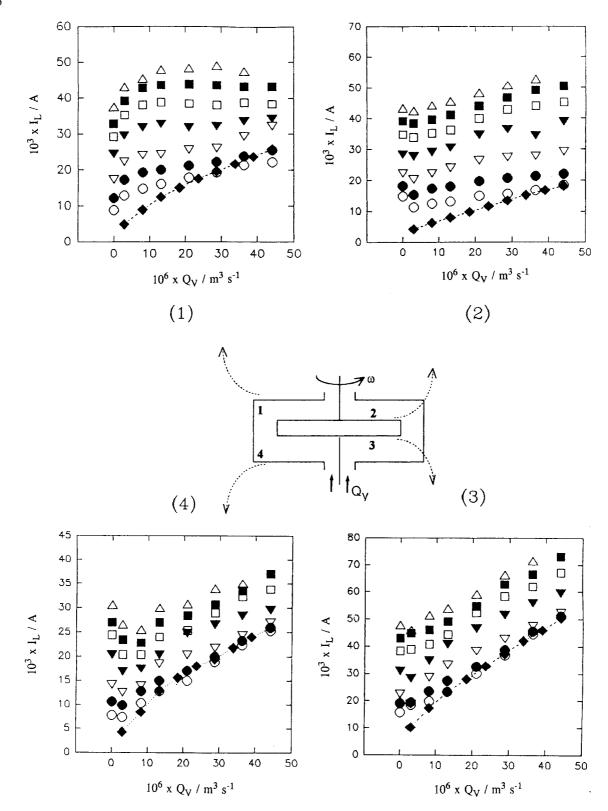
The empirical mass transfer correlations corresponding to exchange surfaces (in bold), and the domains of validity of the correlations, are summarized in Table 1. The lack of lateral confinement in the systems employed by the authors of these correlations, provided interest for experimental work presented here.

Table 1. Empirical correlations for v	Table 1. Empirical correlations for wall-to-liquid mass transfer (combined flow rate and rotation)			
Authors	Correlation	Domain of validity	System and surface	Method
Kreith et al. [14]	$\overline{Sh} = \left(\frac{h}{R_2}\right)^{0.55} \begin{bmatrix} 1.36 + 1.29 \times 10^{-5}Re_{\rm R} + 3.57 \times 10^{-10}Re_{\rm R}^2\\ -3.51 \times 10^{-15}Re_{\rm R}^3 + 1.84 \times 10^{-20}Re_{\rm R}^4\\ \times \begin{bmatrix} Re_{\rm m}^{[0.83-12\times10^{-4}Re_{\rm R}]} \end{bmatrix}$	Sc = 2.4 $0 < Re_{R} < 4 \times 10^{4}$ $5 \times 10^{3} < Re_{m} < 1 \times 10^{5}$ $0.12 < 2h/R_{2} < 0.6$		Dissolution
Jansson and Ashworth [15]	$\overline{Sh} = 1.56  imes 10^{-3} \left( Re_{ m C} rac{H}{R_2} Re_{ m R} rac{H^2}{R_2^2 - R_1^2}  ight)^{0.6}$	$Sc = 8408 \times 10^{-4} < 2h/R_2 < 9 \times 10^{-3}7 \times 10^5 < Re_{\rm R} < 1.5 \times 10^6$		Electrochemical
	$\overline{Sh} = 5.6 \times 10^{-3} \left( Rec \frac{H}{R_2} ReR \frac{H^2}{R_2^2 - R_1^2} \right)^{0.5}$			
Groroghchian et al. [17]	$\overline{Sh} = 4.9 \left( Re_{\rm R} \frac{\hbar^2}{R_2^2 - R_1^2} \right)^{0.57} Sc^{1/3}$ $\overline{Sh} = 4 \times 10^{-3} Sc^{1/3} Re_{\rm m}^{0.1} Re_{\rm R}^{0.57}$	$\begin{array}{l} 4.7\times10^{4} < Re_{\rm R} < 1.5\times10^{5} \\ 3\times10^{4} < Re_{\rm C} < 13\times10^{4} \\ 2h/R_{2} = 0.025 \\ Re_{\rm R} < 4.7\times10^{4} \end{array}$		Electrochemical
Present work	$\overline{Sh} = 1.19 \ Re_{\rm C}^{0.07} Re_{\rm R}^{0.43} \left(\frac{H^2}{R_2^2 - R_1^2}\right)^{0.35} Sc^{1/3}$	$\begin{array}{l} 395 < Re_{\rm C} < 2.3 \times 10^4 \\ 3.5 \times 10^3 < Re_{\rm R} < 8.7 \times 10^4 \\ 4.6 \times 10^{-3} < 2h/R_2 < 0.074 \end{array}$		Electrochemical
	$\overline{Sh} = 0.35 \ Re_{\rm C}^{0.14} Re_{\rm R}^{0.4} \left(\frac{H^2}{R_2^2 - R_1^2}\right)^{0.38} Sc^{1/3}$			
	$\overline{Sh} = 1.58(Re_CRe_R)^{0.26} \left(\frac{H^2}{R_2^2 - R_1^2}\right)^{0.69} Sc^{1/3}$			
	$\overline{Sh} = 0.2 \left( Re_{\rm C} Re_{\rm R} \frac{H^2}{R_2^2 - R_1^2} \right)^{0.27} Sc^{1/3}$			

Table 1 Empirical correlations for wall-to-liquid mass transfer (combined flow rate and rotation)

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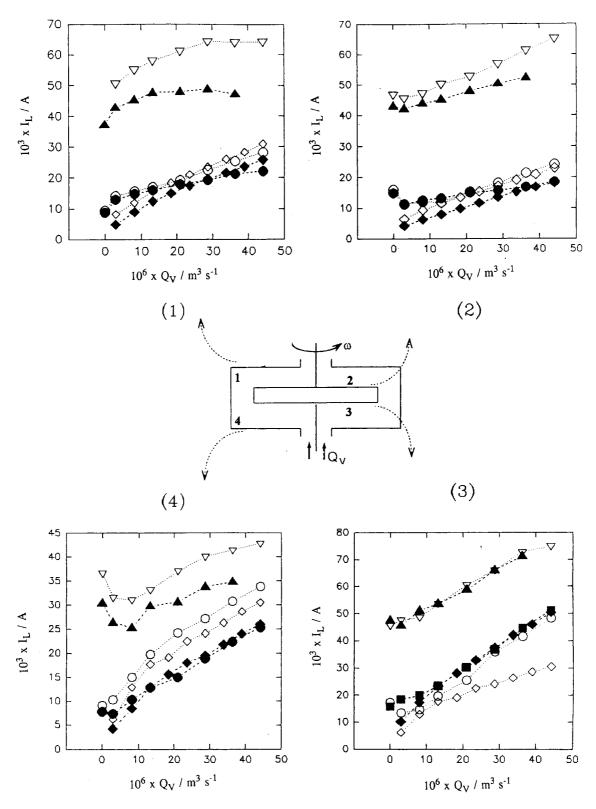


*Fig.* 2. Mass transfer to surfaces *I* to 4: variation of  $I_L$  with  $Q_V$  at constant *N* (for 2h = 8 mm;  $2R_1 = 12$  mm). Key for *N*: ( $\blacklozenge$ ) 0, ( $\bigcirc$ ) 50, ( $\blacklozenge$ ) 100, ( $\bigtriangledown$ ) 200, ( $\blacktriangledown$ ) 400, ( $\square$ ) 600, ( $\blacksquare$ ) 800 and ( $\triangle$ ) 1000 rpm.

The recirculation cell considered in [8] is altered when there is a supporting rotating rod in the space between the two discs [1]. The addition of pumped flow will interfere with rotation and lead to a modified flow structure; mass transfer at surfaces I to 4 could be distinctly affected by the combination of forced flow and rotation.

#### 3. Experimental results

The cell and the geometrical parameters (Figure 1), the experimental method (electrochemical reduction of ferricyanide), and the experimental details, were the same as in Part I [1]. The mean mass transfer coefficient,



*Fig. 3.* Mass transfer to surfaces *I* to *4*: variation of  $I_L$  with  $Q_V$  at constant *N* (for 2h = 4 mm and 8 mm;  $2R_1 = 12 \text{ mm}$ ). Key for *N*: ( $\blacklozenge$ ) 0, ( $\blacksquare$ ) 50 and ( $\blacktriangle$ ) 1000 rpm at 2h = 8 mm; ( $\diamondsuit$ ) 0, ( $\bigcirc$ ) 50 and ( $\bigtriangledown$ ) 1000 rpm at 2h = 4 mm.

 $\bar{k}_{d}$ , was calculated from the measured limiting diffusion current,  $I_{L}$ , according to the following expression:

where *F* is the faradaic constant,  $A_e$  the area of the transfer surface (cathode) and  $C_{\infty}$  the concentration of ferricyanide ions.

In each Figure showing experimental results, a small diagram of the elementary cell indicates the correspon-

$$\bar{k}_{\rm d} = \frac{I_{\rm L}}{FA_{\rm e}C_{\infty}}$$

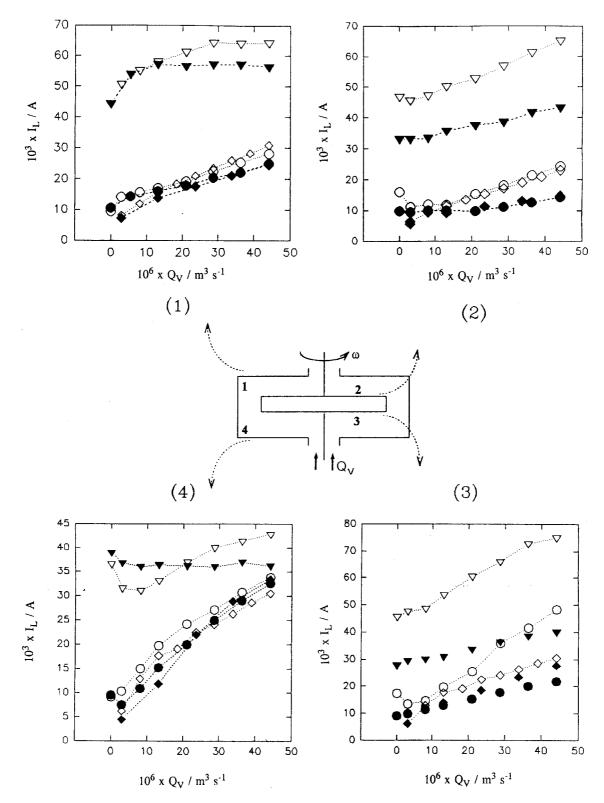


Fig. 4. Mass transfer to surfaces 1 to 4: variation of  $I_L$  with N at constant  $Q_V$  (for 2h = 4 mm;  $2R_1 = 12 \text{ mm}$  and 20 mm). Key for N: ( $\diamond$ ) 0, ( $\bigcirc$ ) 50 and ( $\nabla$ ) 1000 rpm at  $2R_1 = 12 \text{ mm}$ ; ( $\blacklozenge$ ) 0, ( $\blacklozenge$ ) 50 and ( $\nabla$ ) 1000 rpm at  $2R_1 = 20 \text{ mm}$ .

dence between the exchange surfaces and the data presented.

# 3.1. Convergent flow region

# Analysis of Figures 2 to 5 reveals the influence of different parameters (rotation rate N; volumetric flow rate $Q_V$ ; distance 2h; diameter $2R_1$ ) on the limiting diffusion current, $I_L$ , at surfaces I to 4 although the range of parameters as 2h and $2R_1$ is small.

### 3.1.1. Surface 1

 $I_{\rm L}$  increases with N at constant  $Q_{\rm V}$ , but the effect of  $Q_{\rm V}$  is greater at small values of N (N < 200 rpm). This means that mass transfer depends in this domain on both  $Q_{\rm V}$  and N. Beyond N = 200 rpm, mass transfer is controlled

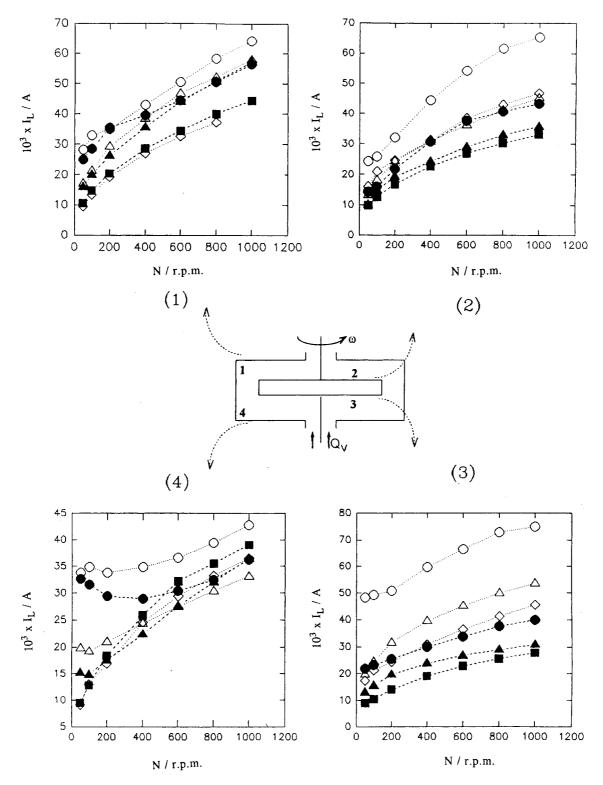


Fig. 5. Mass transfer to surfaces *I* to 4: variation of  $I_L$  with  $Q_V$  at constant *N* (for 2h = 4 mm;  $2R_1 = 12$  mm and 20 mm). Key for  $Q_V$ : ( $\diamond$ ) 0, ( $\diamond$ ) 13 × 10<sup>-6</sup> and ( $\bigcirc$ ) 44 × 10<sup>-6</sup> m<sup>3</sup> s<sup>-1</sup> at  $2R_1 = 12$  mm; ( $\blacksquare$ ) 0, ( $\blacktriangle$ ) 13 × 10<sup>-6</sup> and ( $\bigcirc$ ) 44 × 10<sup>-6</sup> m<sup>3</sup> s<sup>-1</sup> at  $2R_1 = 20$  mm.

by rotation only ( $I_L$  becomes rapidly independent of  $Q_V$  as  $Q_V$  is increased from zero). Figure 3 shows the variation of  $I_L$  with  $Q_V$  at different values of N for two interdisc gaps: all the other parameters being constant, a decrease in 2h improves the transfer. As  $2R_1$  was only varied from 12 to 20 mm, a conclusion cannot be drawn concerning its influence (Figures 4 and 5), except that at

high  $Q_V$  values there is a mass transfer improvement when  $2R_1$  decreases.

#### 3.1.2. Surface 2

 $I_{\rm L}$  increases with N and with  $Q_{\rm V}$  (Figure 2); the increase is small when 2h decreases (Figure 3) whereas a decrease in  $2R_1$  leads to an appreciable improvement in  $I_{\rm L}$  (Figures 4 and 5). As the respective influences of N,  $Q_V$ , 2h and  $2R_1$  on  $I_L$  appear to be simple, it can be expected that empirical correlations including these parameters would be easily obtained.

#### 3.2. Divergent flow region

#### 3.2.1. Surface 3

At a given value of N, the influence of  $Q_V$  on  $I_L$  is more pronounced than for surface 2 (Figure 2). For N < 200 rpm, the experimental points are few but this domain may correspond to a dominant effect of  $Q_V$ . Figures 3 and 4 show that a change of 2h from 4 to 8 mm results in mass transfer improvement, especially at small values of N. At a given  $Q_V$ , there is a decrease of  $I_L$ when  $2R_1$  is changed from 12 to 20 mm, and the effect on  $I_L$  is more marked as N is high. As indicated above, the observed effects of N,  $Q_V$  and  $2R_1$  suggest that empirical correlations including these parameters would be easily obtained.

#### 3.2.2. Surface 4

 $I_{\rm L}$  increases with N at constant  $Q_{\rm V}$  (Figure 2). However, at constant N,  $I_{\rm L}$  first decreases and then increases when  $Q_{\rm V}$  increases from zero. In other words, the experimental curves  $I_{\rm L}$  against  $Q_{\rm V}$  at constant N present two domains (Figure 3): (i) at low values of  $Q_{\rm V}$  [ $Q_{\rm V} < 10 \ (\mu {\rm m})^3 \, {\rm s}^{-1}$ ], where  $I_{\rm L}$  decreases when  $Q_{\rm V}$  increases; and (ii) at higher values of  $Q_{\rm V}$ , where  $I_{\rm L}$  increases with  $Q_{\rm V}$ .

By introducing pumped flow progressively, first the organized structure of the recirculation cells is modified near the entrance of hydraulic diameter 2e; at higher  $Q_V$  values, the cells are compressed against surface 4.

The presence of a minimum in the variations of  $I_L$  with  $Q_V$  is also observed when one considers the influence of

2*h* (Figure 3) and that of the entrance diameter  $2R_1$  (Figures 4 and 5). The value of  $I_L$  obviously results from a competitive influence of  $Q_V$  and N: at low rotation velocities, the influence of  $Q_V$  is dominant while at high rotation velocities N determines the transfer.

Figure 6(a, b) shows the suggested structures in the divergent and the convergent flow regions respectively, while Figure 6(c) reproduces the simple circulating cell verified in [8].

#### 4. Discussion

# 4.1. *Transfer at the four surfaces: qualitative comparison*

For surfaces 1 and 4, the influence of rotation is approximately the same, but the influence of  $Q_V$  is different (progressive control by the rotation for surface 1; combined influence of  $Q_V$  and N for surface 4). The effect of 2h and  $2R_1$  is comparable for both surfaces.

As for the two sides of the rotating disc (surfaces 2 and 3), it appears that: (i) the effect of  $Q_V$  on the mass transfer is higher at surface 3 than at surface 2; (ii) the distance 2h is a parameter for surface 2, not for surface 3; and (iii) at a given  $Q_V$ , the mean liquid velocity in the central annular section increases when  $2R_1$  is decreased and mass transfer is improved.

These results are qualitatively reminiscent of wall to liquid mass transfer at the cylinders of a Couette–Taylor–Poiseuille flow [3, 4] where two domains were distinguished: first, at low flow rates, where the axial flow renders the mean mass transfer coefficient smaller in comparison with rotation alone. Such a decrease in mass transfer is due to eddies stretched in the direction of axial flow; and second, at higher values of  $Q_V$ , where an

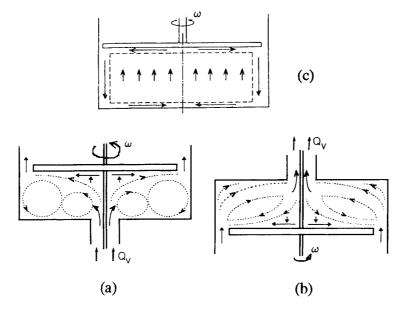
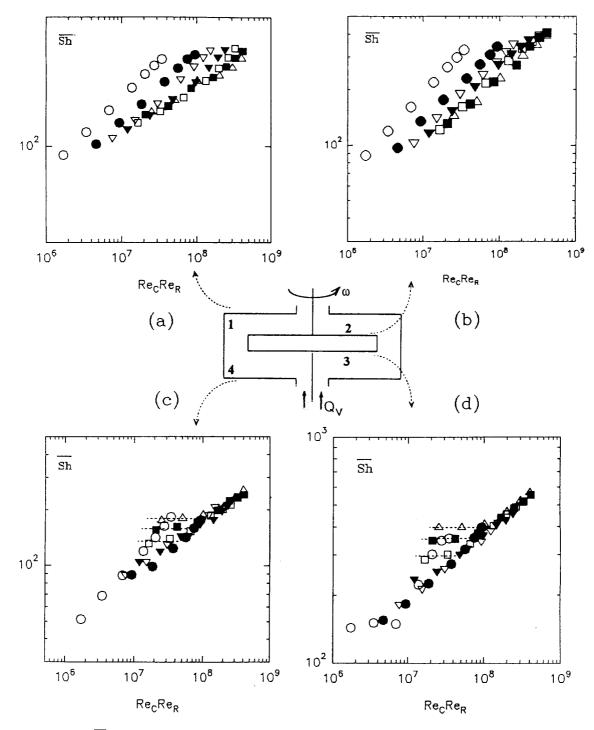


Fig. 6. Suggested flow structures under the combined effect of  $Q_V$  and N (schemes (a) and (b)) and structure below a rotating disc in a box (scheme (c)).



*Fig.* 7. Variations of  $\log(\overline{Sh})$  with  $\log(Re_C \times Re_R)$  for surfaces *l* to *4* (for 2h = 8 mm;  $R_1 = 6$  mm;  $R_2 = 28$  mm).

increase in  $Q_V$  improves mass transfer and is characterized by the presence of eddies of nonuniform size.

For this problem of wall-to-liquid mass transfer in the presence of a Couette–Taylor–Poiseuille flow, empirical correlations proposed for both domains were indicated on an axial Reynolds number against the Taylor number plot.

#### 4.2. Empirical correlations

Ashworth [15, 18] correlated empirically mass transfer results expressing  $\overline{Sh}$  as a dimensionless function:

$$\overline{Sh} = \left(\frac{H}{R_2}\frac{H^2}{R_2^2 - R_1^2}Re_{\rm R}Re_{\rm C}\right)^{\beta}$$

with  $\beta = 0.5$  for the stationary disc and  $\beta = 0.6$  for the rotating disc. Ashworth [15, 18] put in evidence the difficulty to correlate empirically, in a wider domain, mass transfer results corresponding to each disc. For the complex Couette–Taylor–Poiseuille flow, the flow structure is similar at both cylinders and empirical mass transfer correlations can be obtained in extended hydrodynamical flow regimes. In the present case, not only

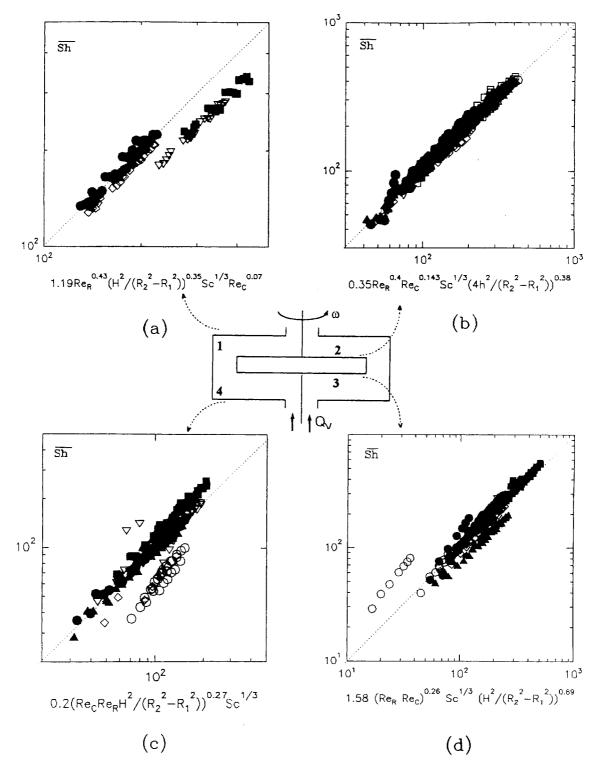


Fig. 8. Empirical correlations for surfaces 1 to 4.

the flow structure is not the same over the different discs but the experimental parameters exert a much stronger influence on structure.

The Rossby number  $Ro = Re_C/Re_R$  permits the distinction between three types of mass transfer control: by forced flow, rotation and mixed control. The current results lead to the following deductions: (i) for Ro < 0.05, mass transfer is controlled by rotation, and the empirical correlation would have the form  $\overline{Sh} \propto f(Re_{\rm R})$ ; (ii) for Ro > 1.3, mass transfer is controlled by forced flow, and  $\overline{Sh} \propto f(Re_{\rm C})$ ; (iii) for 0.05 < Ro < 0.5, mixed control is expected and  $\overline{Sh} \propto f(Re_{\rm C}, Re_{\rm R})$ , that is,  $\overline{Sh} \propto f(Re_{\rm C} \times Re_{\rm R})$  according to the conclusions of Ashworth.

Figure 7 shows bilogarithmic representations of  $\overline{Sh}$  against  $Re_{\rm C} \times Re_{\rm R}$ . For surfaces 3 and 4 (Figures 7(d, c)), each short horizontal dotted line corresponds to a given value of  $Q_{\rm V}$ , and is characteristic of a control

by forced flow; these dotted lines tend to the straight line log  $\overline{Sh} \propto \log(Re_{\rm C} \times Re_{\rm R})$  which characterizes mixed control. No general correlation taking into account the two types of control could readily be established.

If the data corresponding to mixed control are considered, the final empirical correlations in Figure 8 and summarized in Table 1 are obtained. Note in Figure 8(c, d) that a set of points corresponds to  $H/(R_2 - R_1) < 0.1$ , very close discs (H = 2 mm), and a Rossby number higher than 1.3, a value which characterizes control by forced flow.

In these correlations corresponding to surfaces 3 and 4 under mixed control, the exponent of the product  $Re_{\rm R} \times Re_{\rm C}$  is 0.26–0.27, even if hydrodynamic conditions near the surfaces are not exactly the same. In contrast, the exponents of the term  $H^2/(R_2^2 - R_1^2)$  are different in the two correlations, and they differ from those given by Ashworth [15] for the corresponding terms (Table 1), probably because the hydrodynamic domains explored in [15] and in the present work are different.

In the presence of forced flow only (N = 0), it was well established empirically in Part I [1] that mass transfer to surface 3 varies as  $Q_V^{0.62}$  and not  $Q_V^{0.5}$  similar to mass transfer at a disk impinged by a jet [19]. The results shown in Figure 4 for surface 3 and N = 0establish the influence of the flow rate as  $Q_V^{0.57}$  and  $Q_V^{0.55}$ , respectively, for the two values of  $2R_1$ . However, comparison with jet impingement is not fully justified, since the entering jet was not only confined but it was also formed around a rod, and it was concluded in [1] that these two particular factors lead to mass transfer improvement.

At surfaces *I* and *2* mass transfer is mainly controlled by rotation. This is well confirmed by the graphical correlations (Figures 8a, b) and by comparison of the exponents of  $Re_R$  and  $Re_C$  in each correlation shown in Table 1.

At a given value of the product  $Re_R \times Re_C$ , *Sh* is smaller for surface 4 than for the other three surfaces (Figure 7). This means that limiting mass (or heat) transfer, which has to be considered in the design of a cell depicted by Figure 1, occurs at surface 4. Unfortunately, owing to the great variation of the exponents in the correlations of Table 1, a graphical comparison of all the correlations cannot be made.

#### 5. Conclusions

Mass transfer between a liquid and disks in a system combining pumped flow and rotation of a category of discs, presents a complex problem which depends on geometrical and hydrodynamic parameters (i.e., flowrate, rotation velocity). Only an empirical analysis of the results is possible, as in the extreme cases of forced flow only and rotation only considered in Part I [1]. At given experimental conditions, the smaller value of the mass transfer coefficient corresponds to the stationary surface (surface 1) situated in the convergent flow region. This is important because it indicates that surface 1 is mass transfer limiting in the entire cell. In the case of an electrochemical reactor, this also means that the maximum current density to be applied in the cell would be the limiting current density at surface 1. In the convergent flow region, mass transfer is controlled by rotation while the control is mixed in the divergent flow region. An empirical correlation was established for each of the four surfaces.

Mass transfer studies using microelectrodes in an insulated or conducting wall offer a useful perspective for the investigation of pertinent hydrodynamic flow structures.

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