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Entrance effect on mass transfer in a parallel plate electrochemical reactor

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Abstract

The influence of different fluid inlet types, slits or tubes, on mass transfer in a rectangular reactor was studied. Measurements of mass transfer coefficients were made using the limiting diffusion current technique based on ferricyanide ion reduction at a large nickel electrode located downstream of abrupt expansions. The overall mass transfer coefficients obtained were 3 to 13 times greater than those obtained in fully developed flows. Overall mass transfer coefficients were correlated for Reynolds numbers ranging from 400 to 3500 by a unique equation by introducing a nondimensional expansion factor defined by the ratio of the fluid inlet cross-section to that of the reactor. The correlation equation obtained was compared with literature data.

List of symbols

D	diffusion coefficient	s	cross-sectional area of the fluid inlet
D_e	equivalent diameter of the electrochemical cell	Sc	Schmidt number ($= v/D$)
d_e	equivalent diameter of the entry arrangement	Sh	Sherwood number ($= KD_e/D$)
h	step height	Sh_{FD}	Sherwood number in fully developed flow
K	overall mass transfer coefficient	Sh_{max}	peak Sherwood number
L	electrode length	U	mean axial velocity in the electrochemical cell
Re	Reynolds number ($= UD_e/v$)	v	kinematic viscosity
S	cross-sectional area of the electrochemical cell		

1. Introduction

Parallel plate-type cells are very often used as electrochemical reactors [1–5]. Entrance devices have an important influence on the performance of such reactors which generally operate with developing flows. The entrance region is characterized by the presence of recirculating flow generated downstream of the inlet expansion. Much work has been devoted to the study of heat or mass transfer in sudden expansion flows and significant enhancement of heat/mass transfer is observed. Local heat transfer coefficients were determined [6–8] in turbulent flow with constant heat flux condition in cylindrical sudden expansions of different expansion ratios. Filetti and Kays [9] introduced an expansion factor to correlate the peak heat transfer coefficient in turbulent sudden expansion flow between parallel plates. The electrochemical method is generally used to study the mass transfer in such configurations. Runchal [10] has obtained the local mass transfer coefficients in cylindrical expansion flow with an expansion factor of 0.5, for Reynolds and Schmidt numbers respectively comprised between 2550 and 88 500, and 1400 and 2500. Tagg et al. [11] have investigated the effect of the expansion factor on mass transfer. The mass transfer distribution in the recirculation zone is a function of the Reynolds number and the expansion factor. Wragg et al. [12] have analysed the mass transfer in two types of expansion flows: a tube pipe expansion and the expansion of a cylindrical tube into a square duct. The mass transfer behaviour is similar in both cases in the axial direction, but a lateral variation of mass transfer is observed for the expansion in square duct. However, if the Reynolds number is defined using the diameter of the entrance tube as characteristic length, the exponent of the Reynolds number is similar in the different correlations. Recently, Rizk et al. [13] showed that the mass transfer enhancement after sudden

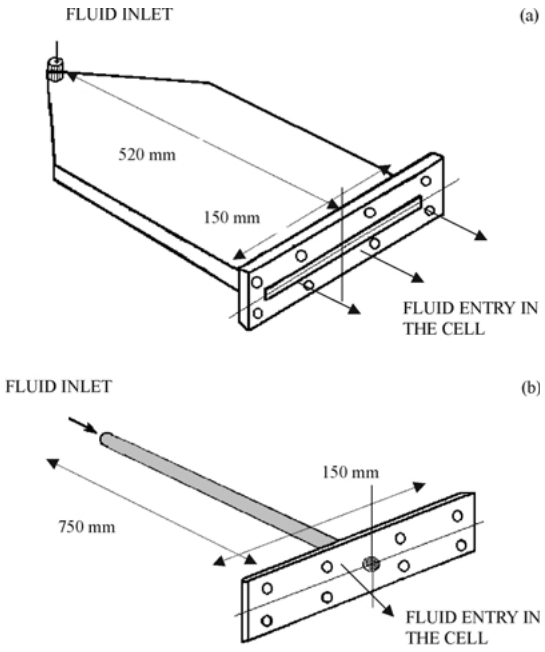

Fig. 1. Fluid injection arrangement: (a) slit-type entry, (b) tube-type entry.

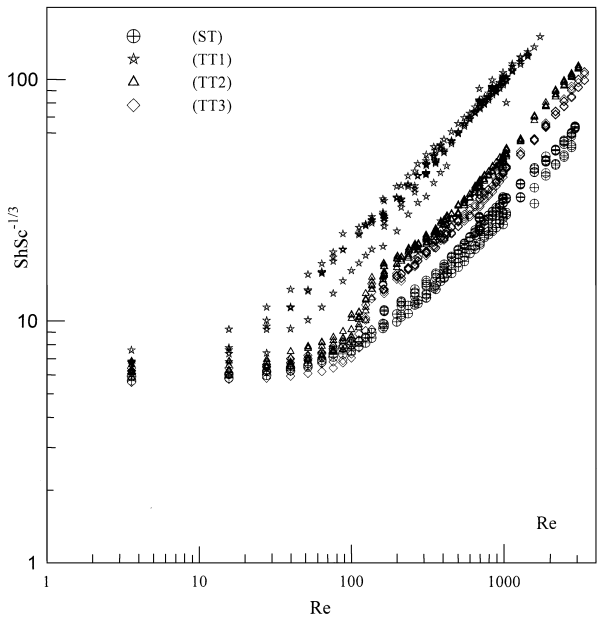

Fig. 2. Variation of $Sh\ Sc^{-1/3}$ with *Re*.

expansion is inversely proportional to the Reynolds number.

The aim of this work is to study the effect of the type of fluid entry in a parallel plate reactor. The mass transfer coefficients are determined by an electrochemical method downstream of different fluid entry arrangements in the case of simultaneous development of mass and momentum boundary layers. A dimensionless expansion factor is proposed to take the different types of expansion flow into account in order to establish a unique correlation for all experimental data.

2. Experimental details

The experimental cell consisted of a rectangular duct 150 mm wide, 20 mm high and 500 mm long. Two 150 mm × 300 mm nickel electrodes were set up at the entrance of the electrochemical cell. Two types of fluid entrance were studied (Figure 1): a slit-type entrance (ST) 150 mm wide and 2 mm high and a tube-type entrance (TT) with three different diameters. The different fluid inlets are characterized by their hydraulic diameter and by the ratio s/S of the cross sectional area of the inlet to that of the electrochemical cell (Table 1). The effect of s/S on mass transfer was studied in the domain 9.4×10^{-3} (for TT1) $< s/S <$ 0.1 (for ST). The flow regime was established in the slit and tubes before the fluid entered the electrochemical cell. The entrance systems have been symmetrically set up with respect to the electrochemical cell at the cell inlet (Figure 1). The rig carrying the electrochemical cell consisted of a reservoir with a temperature control device, a centrifugal pump and two rotameters covering a flow rate range 1–800 $dm^3\ h^{-1}$. The mass transfer coefficients on the nickel electrodes were obtained by using the well-known technique based on the determination of diffusional limiting current by performing the cathodic reduction of ferricyanide ion. The working solution consisted of 2×10^{-3} M potassium ferricyanide, 5×10^{2} M potassium ferrocyanide and 0.5 M sodium hydroxide, as supporting electrolyte. The experiments were conducted at 30 °C, the kinematic viscosity was $8.73 \times 10^{-3}\ cm^2\ s^{-1}$, the diffusion coefficient of ferricyanide ion was $6.95 \times 10^{-6}\ cm^2\ s^{-1}$ and the Schmidt number was 1260.

3. Results and discussion

Mass transfer is expressed in terms of $Sh \times Sc^{-1/3}$ as a function of the Reynolds number *Re* defined using the characteristics of the cell. The mass transfer data for the four different entrance systems are given in Figure 2. Two flow regimes are observed. For low values of the Reynolds number, the mass transfer regime is characterized by constant mass transfer coefficients, independent of Reynolds number. Mass transfer results in the

Table 1. Geometric characteristics of the entry arrangement

Type of entrance system	Slit-type (ST)	Tube-type (TT1)	Tube-type (TT2)	Tube-type (TT3)
d_e/mm	3.95	6	12	15
s/S	0.1	9.4×10^{-3}	3.77×10^{-2}	5.89×10^{-2}

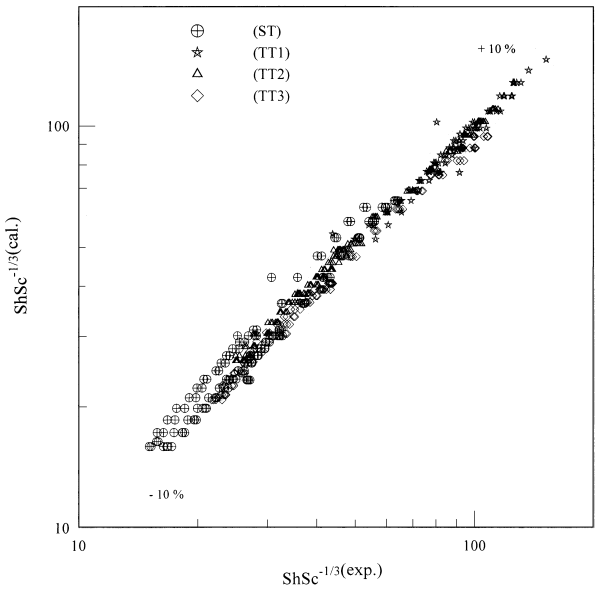

Fig. 3. Comparison between experimental data with Equation: 6 $Sh\ Sc^{-1/3} = 0.068\ Re^{0.72}(s/S)^{-0.5}$.

competition between different mechanisms associated with the two flow configurations: the set-up, just downstream of the flow expansion, of a recirculation bubble and the development of the laminar boundary layer at the second part of the electrode. For low values of Reynolds number ($Re \leqslant 10$), the recirculation bubble length is very small [14] and the position of the reattachment point ranges from 0 to 2 h, where h is the step height. For laminar flow, the position of the reattachment point increases sharply with Re, up to $Re = 400$–500 [14, 15]. Thus, the weak variation of the recirculation bubble size for low Reynolds number leads to quasi-constant mass transfer. The constant value of $Sh\ Sc^{-1/3}$ is equal to 6–8 and seems to be independent of the fluid entrance type. In contrast, the value of the critical Reynolds number, corresponding to transition between the two flow regimes, is a function of the entry arrangement (Figure 3). The second flow regime is characterized by a sharp change of $Sh\ Sc^{-1/3}$ for Reynolds number values greater than 200–400 (Figure 2). For each entrance system, the following correlations are obtained:

$$(\mathrm{ST})\ Sh\,Sc^{-1/3} = 0.30\,Re^{0.66} \tag{1}$$

$$(\mathrm{TT1})\ Sh\,Sc^{-1/3} = 0.65\,Re^{0.73} \tag{2}$$

$$(\mathrm{TT2})\ Sh\,Sc^{-1/3} = 0.31\,Re^{0.73} \tag{3}$$

$$(\mathrm{TT3})\ Sh\,Sc^{-1/3} = 0.26\,Re^{0.73} \tag{4}$$

The mass transfer coefficients increase with decrease in the equivalent diameter of the tube-type (TT) entrance (see Table 1 for a description of the different TT-type fluid entrances). A slit-type (ST) entrance leads to lower mass transfer coefficients compared with tube-type entrances. The value of the exponent of Reynolds number is quasi-similar for both types of entrance and very close to that obtained in fully turbulent channel flow. The Reynolds number domain characterizing the transition zone between the two flow regimes shifts toward lower values of Re with increase of the mass transfer level, that is, when the entrance equivalent diameter decreases. The first attempt to correlate the mass transfer coefficients obtained with the different entry arrangements is to introduce the ratio d_e/D_e of the entrance equivalent diameter to the electrochemical cell equivalent diameter. This parameter has been used to express the peak heat/mass transfer [7, 11] in axisymmetric sudden expansion. The equation:

$$Sh\,Sc^{-1/3} = 0.11\,Re^{0.73}\left(\frac{d_e}{D_e}\right)^{-1} \tag{5}$$

accurately correlates the mass transfer data obtained with the three tube-type inlets. However, Equation 5 cannot be used for data obtained with the slit-type entrance, which leads to the lowest mass transfer with the lowest equivalent diameter. For this reason, we have proposed a new expansion factor s/S, defined by the ratio of entrance cross-section area to that of the cell. This factor takes two geometric dimensions into account, which may be pertinent for the slit-type entrance. All experimental data for Reynolds number values between 400 and 3500 are correlated by

$$Sh\,Sc^{-1/3} = 0.068\,Re^{0.72}\left(\frac{s}{S}\right)^{-0.50} \tag{6}$$

The comparison of Equation 6 with the experimental data is given in Figure 3. The agreement is good and a deviation of 8% is found. Thus, the expansion factor s/S seems to be a reliable parameter to characterize the flow expansion. It also corresponds to the ratio of the mean velocity in the electrochemical cell to that one in the fluid entrance. The mass transfer is proportional to $(s/S)^{1/2}$. This result leads to consideration of $(s)^{1/2}$ as a mass transfer characteristic length.

Wragg et al. [12] have studied the peak mass transfer in tube-type expansion flow in tube and square channels. They have proposed the following expression:

$$Sh_{\max}\,Sc^{-1/3} = 0.27\,Re^{0.67}\left(\frac{d_e}{D_e}\right)^{-0.67} \tag{7}$$

for the expansion into a tube. Equation 7 can be rewritten as

$$Sh_{\max}\,Sc^{-1/3} = 0.27\,Re^{0.67}\left(\frac{s}{S}\right)^{-0.335} \tag{8}$$

and for the expansion into the square channel as

$$Sh_{\max}\,Sc^{-1/3} = 0.29\,Re^{0.67}\left(\frac{s}{S}\right)^{-0.335} \tag{9}$$

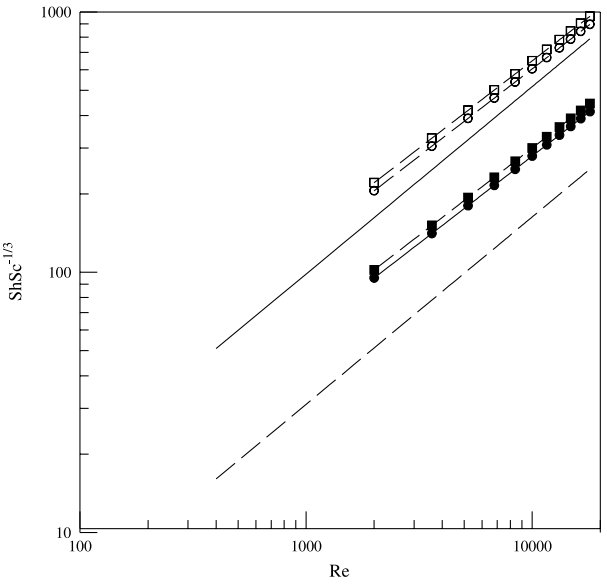

Fig. 4. Comparison of Equation 7 with the peak mass transfer in expansion flows obtained by Wragg et al. [12]. Key: (□, ■) expansion tube–tube; (○, ●) expansion tube-square. Sources: (■, ●) Wragg et al. [12], $s/S = 0.1$; (- - - -) present study, $s/S = 0.1$; (□, ○) Wragg et al. [12], $s/S = 0.01$; (—) present $s/S = 0.01$.

It can be observed in Figure 4 that the peak mass transfer coefficients obtained by Wragg et al. [12] are logically greater than those calculated by Equation 6, which are determined from global mass transfer measurements. In both cases, the mass transfer increases with decrease in the expansion factor s/S which corresponds to an increase of the recirculation flow downstream of the sudden expansion. It is also observed in Figure 4 that the difference between the two sets of data is less important when s/S increases. The axial extension of the recirculation zone with decrease in s/S leads to an enhancement in the mean Sherwood number. The mass transfer in expansion flows is characterized [16] by the separation, reattachment and redevelopment of boundary layers experienced by the flow. The recirculation flow persists up to the reattachment of the separated flow to the wall. The position of the reattachment point is important for mass transfer performance. Pack et al. [14] and Armaly et al. [15] have shown that the reattachment length in symmetrical sudden-expansion pipe flow [14] and in a backward-facing step flow [15] increases with Re in laminar flow for Re between 10 and 200 after Pak et al. [14] and for Re less than 500 after Armaly et al. [15], decreases with Re for transitional regime for Re = 200–800 [14] and for Re = 500–2000 [15] and becomes constant in the turbulent flow regime. In turbulent flows, the abscissa of the reattachment point is equal to 8 h [15], where h is the step height. By assuming that the slit-type (ST) entrance is similar to a backward-facing step, the recirculation zone length in our operating conditions should be equal to 80 mm. For abrupt nozzle expansions in turbulent pipe flow, Tagg et al. [11] have shown that the peak mass transfer is obtained inside the recirculation bubble and that the recirculation zone length increases with a decrease in d_e/D_e greater than 1/4. They have also observed a slight decrease in the recirculation zone length for Reynolds numbers between 4000 and 24 000. Under the experimental conditions of [11], the recirculation zone length lies within 3 and 4 D_e. For Re ranging from 1000 to 10 000, Pak et al. [14] have obtained larger reattachment lengths. For $d_e/D_e = 3/8$, the reattachment length is constant and lies between 6 and 10 taking account of the experimental uncertainty. By using these results in our experimental conditions, the recirculation zone length would be between 100 and 350 mm. This means that the recirculation bubble is bigger for a tube-type entrance than for a slit-type. Thus, the difference in mass transfer between the two entrance systems can be explained by the recirculation bubble size and by the fluid velocity injection in the electrochemical cell, which is related to the expansion factor s/S.

To evaluate the mass transfer enhancement with respect to the fully developed turbulent channel, Equation 6 is rewritten as

$$Sh\,Sh_{FD}^{-1} = 1 + 0.44\,Re^{+0.066}\left(\frac{s}{S}\right)^{-0.6} \tag{10}$$

Mass transfer in fully developed turbulent channel flow has been determined by Pickett and Ong [17] in a rectangular electrochemical cell for $Re > 4000$ and for L/D_e between 0.17 and 10.1, where L is the length of the mass transfer area. In our experimental conditions ($L/D_e = 8.5$), the equation given by [17] becomes

$$Sh_{FD}\,Sc^{-1/3} = 0.081\,Re^{2/3} \tag{11}$$

By setting $s/S = 1$ in Equation 6, the difference in mass transfer given by Equations 6 and 11 is about 20%. This result emphasizes the reliability of Equation 6 to predict mass transfer coefficients in channel electrochemical cells for different fluid entrance conditions. Depending on the fluid entrance, the mass transfer enhancement is very significant and varies between 3 and 13. It is also noticed that the enhancement increases slightly with Re.

4. Conclusion

Determination of mass transfer in different types of flow expansion has shown that the expansion factor s/S corresponding to the ratio of flow cross-section area in the entrance to that in the cell, is the pertinent parameter to predict mass transfer coefficients. The expansion factor is, in fact, related to the ratio of the velocities in the entrance and in the cell. For both entrance types studied, mass transfer increases with decrease in the expansion ratio. All the experimental data were correlated using a unique equation which is in good agreement with available literature correlations for peak mass transfer in expansion flows. The use of an expansion-type entrance

leads to significant enhancement in mass transfer in the entire electrochemical cell.

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