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The effect of a perpendicular and cumulative inlet flow on the mass-transfer distribution in parallel-plate electrochemical reactors

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ABSTRACT

A perpendicular inlet flow of electrolyte is analysed as a means of improving mass-transfer in a parallel-plate electrochemical reactor. Experimental local Sherwood numbers along the electrode length are reported for different values of the hydraulic diameter and Reynolds number, using the reduction of ferricyanide as a test reaction. The Reynolds numbers, evaluated with the inlet velocity in the reactor, range from 0.7 to 30 with interelectrode gaps varying between 3.2 to 9.8 mm. The perpendicular and cumulative flow makes uniform the mass-transfer distribution at the reactor inlet under laminar flow conditions. The experimental data are compared with theoretical calculations obtained by computational fluid dynamics and also with an analytical simplified model. The mean relative deviation between the experimental and theoretical results was lower than 5% and a close agreement was observed between both theoretical treatments.

1. Introduction

In the industrial practice the mass-transfer has a crucial influence on the performance of the equipments. The improvement of the efficiency of electrochemical reactors requires to increase the mean mass-transfer coefficient and also to become more uniform the mass-transfer distribution along the electrode length. Additionally, in the case of parallel-plate electrodes the behaviour is affected by the entrance and exit effects, which involves an appropriate design of the ports for the inlet and outlet of the electrolyte to the reactor [1]. To address the above issues different alternatives are proposed: (i) placing obstacles in the interelectrode gap to promote turbulence [2], increasing the roughness of the electrode [3] and (iii) sparging a gas in the interelectrode gap or its evolution at the electrode surface [4], which were previously discussed [5]. Other strategy is the modification of specific hydrodynamic aspects of the reactor. Thus, several studies have been devoted to the investigation of different patterns for the flow of the electrolyte inside the equipment. The effect of the geometrical parameters of multijets of electrolyte on the overall mass-transfer behaviour at a flat circular electrode was investigated and a satisfactory empirical correlation of the results was reported [6,7]. Oren et al. [8] analysed the interaction over a flat electrode of two identical solutions jets entering an electrochemical reactor in opposite directions, on the same axis, and leaving it in a direction perpendicular to the inlet flow. This concept, also called impinging streams, has shown only a slight improvement of the mean mass-transfer coefficient. However, the dimensionless ratio of the mass-transfer coefficient to the energy consumption exhibits a better performance in comparison with the single stream mode. Reade et al. [9] studied the effect of a jet flow of electrolyte on the performance of a reticulated vitreous carbon rotating cylinder electrode. The electrolyte jet impinged on the bottom central part of the electrode in line with the rotation axis producing an enhancement in the mass-transfer coefficient between 3–46% depending on the rotation speed and type of electrode material.

Legentilhomme and Legrand [10] studied the mass-transfer characteristics at the inner electrode of an annulus in laminar and turbulent swirling flows induced by means of a tangential inlet in the annular gap. Enhancement of the mean mass-transfer coefficient up to 400% was achieved in comparison to that obtained in fully developed axial flow. This concept was also implemented for the cathodic removal of metal from dilute process solutions [11]. Recently, Martínez-Delgadillo et al. [12] evaluated, using computational fluid dynamics, three different inlet types of a tubular electrochemical reactor and it was found that axial velocity distribution is more homogeneous when the reactor is operated with a tangential inlet.

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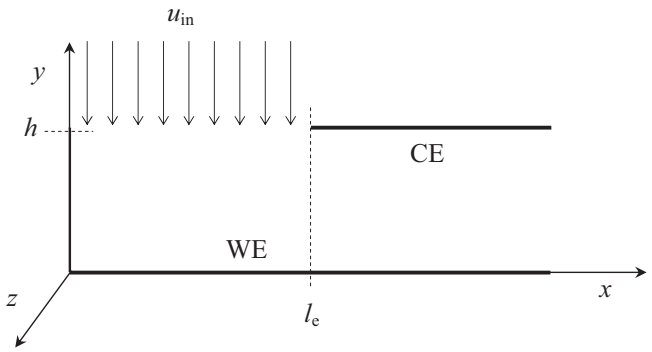

Fig. 1. Schematic view of the reactor with perpendicular inlet flow and coordinates. WE, working electrode; CE, counter electrode.

Cœuret and Legrand [13] proposed the 'falling-film cell', a parallel-plate electrochemical reactor where a gravity flow of electrolyte occurs over an inclined plate and below a sheet of expanded metal positioned above and parallel to the plate. At low flow rates capillary effects are present producing an increase in the mass-transfer coefficient when the flow rate is decreased, called capillary flow regime, which was industrially implemented for the removal of metal ions from effluents.

The use of a pulsed flow reactor was analysed to enhance mass-transfer at the electrode [14], pulsation induces stirring of the electrolyte which is easily controlled by the amplitude and frequency of the pulse. This concept was primarily developed at commercial scale for metal recovery in wastewaters and also for electroorganic synthesis.

In a previous paper from this laboratory [5], a continuous reduction in the cross-section area was analysed as a means of improving mass-transfer in a parallel-plate electrochemical reactor. Thus, it was achieved a convergent flow along the reactor, which improved the mass-transfer coefficient by 10-60% and the mass-transfer distribution under laminar flow conditions was more uniform.

Jorne [15] and Lessner and Newman [16] investigated a cumulative flow channel in which the electrolyte was uniformly fed through a porous wall facing the active electrode. Because of the cumulative nature of the flow, the average velocity increases linearly along the length of the electrode and the diffusion boundary layer becomes uniform. This arrangement is a unique flow system that exhibits uniform accessibility to mass transfer. The concept was applied to the case of a zinc-chloride battery [17]. However, its implementation is difficult in the case of parallel-plate electrochemical reactors when both electrodes are massive.

The aim of this paper is to study in electrochemical reactors with parallel-plate electrodes the effect of a perpendicular inlet flow on the mass-transfer distribution. Thus, the fluid is fed uniformly through a port facing the electrode. The port has the same width as the electrode and the end of the channel is closed off. Because of the cumulative nature of the flow the average velocity increases linearly along the length of the port and the electrode is uniformly accessible to mass-transfer.

2. Mathematical modelling of the mass transfer

Figure 1 sketches the modelled electrochemical system, consisting of a rectangular flow channel where one wall is the working electrode facing the counter electrode that covers partially the opposite wall. Thus, at $y = h$ for $0 \le x \le l_e$ the electrolyte is uniformly fed and in this region the flow is cumulative. For a parallel-plate electrochemical reactor the distribution of the local Sherwood number, Sh_x, along the electrode length is given by [18]

$$Sh_x = \frac{\sqrt{\varepsilon}}{\Gamma(4/3)}\left(\frac{Re_{in}Sc}{9\int_0^X \sqrt{\varepsilon}dX}\right)^{1/3} \quad (1)$$

being

$$\varepsilon = \left.\frac{\partial U_X}{\partial Y}\right|_{Y=0} \quad (2)$$

$$U_X = \frac{u_x}{u_{in}} \quad (3)$$

$$X = \frac{x}{d_h} \quad (4)$$

and

$$Y = \frac{y}{d_h} \quad (5)$$

where the local Sherwood number, Sh_x, the Reynolds number, Re_{in}, and the Schmidt number, Sc, are defined as

$$Sh_x = \frac{k_{m,x}d_h}{D} \quad (6)$$

$$Re_{in} = \frac{u_{in}d_h}{\nu} \quad (7)$$

and

$$Sc = \frac{\nu}{D} \quad (8)$$

here $k_{m,x}$ is the local mass-transfer coefficient, d_h is the hydraulic diameter, ν is the kinematic viscosity, D is the diffusion coefficient and u_{in} is the average fluid velocity at the reactor inlet.

The hydrodynamic regime inside the reactor in steady-state is given by the Navier-Stokes equations

$$U_X\frac{\partial U_X}{\partial X} + U_Y\frac{\partial U_X}{\partial Y} = -\frac{\partial P}{\partial X} + \frac{1}{Re_{in}}\left[\frac{\partial^2 U_X}{\partial X^2} + \frac{\partial^2 U_X}{\partial Y^2}\right] \quad (9)$$

$$U_X\frac{\partial U_Y}{\partial X} + U_Y\frac{\partial U_Y}{\partial Y} = -\frac{\partial P}{\partial Y} + \frac{1}{Re_{in}}\left[\frac{\partial^2 U_Y}{\partial X^2} + \frac{\partial^2 U_Y}{\partial Y^2}\right] \quad (10)$$

Thus, to calculate the local Sherwood number it is necessary to introduce into Eq. (1) the function ε, which requires the simultaneous solution of Eqs. (9) and (10) together with the continuity equation

$$\frac{\partial U_X}{\partial X} + \frac{\partial U_Y}{\partial Y} = 0 \quad (11)$$

being

$$U_Y = \frac{u_y}{u_{in}} \quad (12)$$

where u_x and u_y are the velocities of fluid along the axial coordinates and P is the dimensionless pressure. The variation of the local Sherwood number along the electrode width can be disregarded due to the small ratio between the interelectrode gap and the electrode width [5,19].

Assuming that at the entrance region the velocity component in the y direction is not a function of x, that is

$$U_Y = -\Theta(Y) \quad (13)$$

Eqs. (9-11) are simplified to the following ordinary differential equation [15,16,20]

$$\frac{d\Theta}{dY}\frac{d^2\Theta}{dY^2} - \Theta\frac{d^3\Theta}{dY^3} = \frac{1}{Re}\frac{d^4\Theta}{dY^4} \quad (14)$$