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Local mass-transfer study in a decaying swirling flow electrochemical reactor under single-phase and two-phase (gas-liquid) flow

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HIGHLIGHTS

- The swirling flow improves the mass-transfer conditions in a reactor.
- The swirling flow becomes more uniform the mass-transfer distribution.
- The gas phase has scarce influence on the mass-transfer performance.

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ABSTRACT

Mass transfer was studied at the external electrode of a cylindrical electrochemical reactor with decaying swirling flow using a single phase electrolyte and also a dispersion of a gas phase in the solution. The local mass-transfer coefficients were measured with a segmented cathode using the reduction of ferricyanide as test reaction. Two strategies were used to introduce the gas phase. In the first case the gas was fed by three holes symmetrically drilled at the bottom of the central anode tube and in the second one by means of a tee fitting at the reactor inlet. The best performance was obtained with the single phase electrolyte giving 0.78 as the exponent of the Reynolds number and the mass-transfer distribution along the reactor length is more uniform than that reported for a parallel plate electrochemical reactor under laminar flow conditions. The mass-transfer enhancement factor related to an annular duct with axial developing flow ranges from 8 to 11. This high value is the consequence of two factors: (i) the intensive forced convection produced by the swirling flow at the outer electrode and (ii) the sudden expansion of the flow at the entrance due to the small tangential inlet. The fed of a gas phase in the reactor has scarce influence on the mass-transfer performance. A comparison of mass-transfer results with previous studies is made.

1. Introduction

The enhancement of the space time yield in process equipment is of vital importance in order to increase its performance. Two strategies can be used to improve the space time yield of electrochemical reactors with reactions influenced by mass-transfer. The first is the increase of the specific surface area of the electrode arising three-dimensional electrodes. The second one is the enhancement of the mass-transfer conditions in the reactor by modification of its hydrodynamics. Based on the last concept many techniques have been proposed, being the use of swirling flow a promising one. These equipments present as additional advantages: simple constructive features, absence of moving parts and easy to recover the products. In a pioneer work Cedrone [1] proposed a reactor with tangential solution nozzles, located at the bottom of a cylindrical reactor, in order to improve the mass-transfer for silver deposition onto a external cathode. Along the axial length, the solution continues to swirl and climbs in an ascending helix in the interelectrode gap formed by a concentric tube, working as anode. At the top of the reactor the fluid is deflected to the internal part of the anode where it falls into a filter sock suspended within the anodic tube and it runs downward into a sump tank, being again recirculated. Other application of the concept of helical flow was reported by Walsh and Wilson [2] for the removal of gold from spent metal finishing process liquors by means of the “Metelec Concentric Cylindrical Cell”, which incorporates a cylindrical foil cathode concentrically arranged around an inner anode. The inlet and outlet manifolds were tangentially positioned near the bottom and top of the cell, respectively. However, the concept of swirling flow was scarcely explored in electrochemical

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reactors for effluents treatments from dilute solutions, where the achievement of good mass-transfer conditions is a very important goal. Wang [3,4] proposed, for the electrowinning, electrorefining or recovery of metals, the EMEW cell (EMEW Clean Technology, Vancouver, BC, Canada), which consists of a pair of concentric tubes, the inner being an iridium/ruthenium-coated titanium anode and the outer being a rolled stainless-steel sheet cathode held by a polyvinyl chloride outer body. At both ends of the cell are plastic closure caps with liquor inlet and outlet ports.

Likewise, this rotary type of liquid flow causes a reduction in pressure in its central axial part. Then, a gas phase can be introduced to the reduced pressure area. This concept is described as a procedure for the generation of a gas-liquid dispersion with the formation of microbubbles [5], which can be recognised as an interesting proposal for the electrochemical processing of gaseous reagents.

The foregoing industrial applications were developed simultaneously with studies of mass-transfer in these systems, which are under a special hydrodynamic condition frequently called helical or swirl flow. This flow may be classified into two different types according to the method used to achieve the rotation movement: continuous swirl and decayed or damped swirl. In the first type, the swirling motion persists and maintains its characteristics over the entire length of the tube or annular space, while in the second type, the swirl is generated at the inlet section and decays freely along the flow path and its properties vary with axial distance from the entrance. Continuous swirl is usually achieved by inserting twisted tapes, spiral fins or coiled wires of varying pitch, helix angle and wire diameter, along the entire length of the tube. Decaying swirl may be induced by tangential inlet slots or tubes, tangential vanes, short lengths of helical inserts, and rotating cylinders or propellers. The second type is preferred because of its simple design and easy maintenance, due to the absence of inserts, and also by a lower pressure drop in the equipment. Shoukry and Shemilt [6] reported results of mass-transfer in turbulent decaying swirl flow in an annulus. Enhancement of the global mass transfer coefficient of up to 320% was obtained, in comparison with the value for developed axial flow. Legentilhomme and Legrand [7–9] studied the mass-transfer between a liquid and the inner cylinder of an annular gap under decaying swirling flows and an enhancement factor of the global mass-transfer coefficient up to 4 was reported. de Sa et al. [10] corroborated the improvement of the global mass-transfer coefficients by a decaying swirling flow. Yapici et al. [11–13] reported a study of the local mass-transfer behaviour, measured with microelectrodes, in decaying annular swirl flow produced by axial vane-type generators. The mass-transfer coefficients decay towards the value for fully developed flow as the axial distance increases, but, even at 50 equivalent diameters downstream, the effect of swirl is still evident. Lefèbvre et al. [14] corroborated that the mass-transfer coefficient for the outer wall is higher than that for the inner rod. The comparison with previous studies allowed concluding that the use of single tangential entries, to generate the swirl flow, enhances the mass-transfer coefficient due to the higher swirl intensity. In a further contribution, Yapici et al. [15] studied the local mass-transfer distribution in decaying swirl flow in a circular pipe.

The previous studies on mass-transfer in decaying swirl flow do not provide any detailed information on the local mass-transfer distribution for reactors with cylindrical concentric electrodes. This paper describes the results of experimental work on local mass-transfer measurement in decaying annular swirl flow using an electrochemical technique involving a segmented electrode in a home-made reactor. Both, single phase and biphasic system, gas-liquid, were studied.

2. Material and methods

The reactor, shown schematically in Fig. 1, was made of two parallelepiped blocks of acrylic material, 80 mm side and 149 mm length, which were assembled by a threaded joint with an O'ring seal 1 mm thick. In both blocks, it was machined a cylindrical cavity of 42 mm diameter in whose internal wall the segmented cathode was arranged, made of 20 segments of 316 stainless steel, 38.5 mm internal diameter and 12 mm length. The segments were insulated from one another by a polyamide ring of approximately 1 mm thick. Thus, the total length of the segmented cathode was 257.5 mm and its surface was polished with emery paper 2500 grade. The insulating insertions between segments disturb the concentration profiles at the electrode surface [16] altering the mass-transfer behaviour. However, in a previous work [17] it was demonstrated using computational fluid dynamics that the maximum error in the measurement of the mass-transfer coefficient, as a result of the use of a segmented electrode with level separators, may be in the order of 14%, which is an acceptable value for this type of experiments. However, the presence of recessed insulating insertions between segments additionally produces a turbulence promotion action generating a greater increase in the mass-transfer coefficient [18].

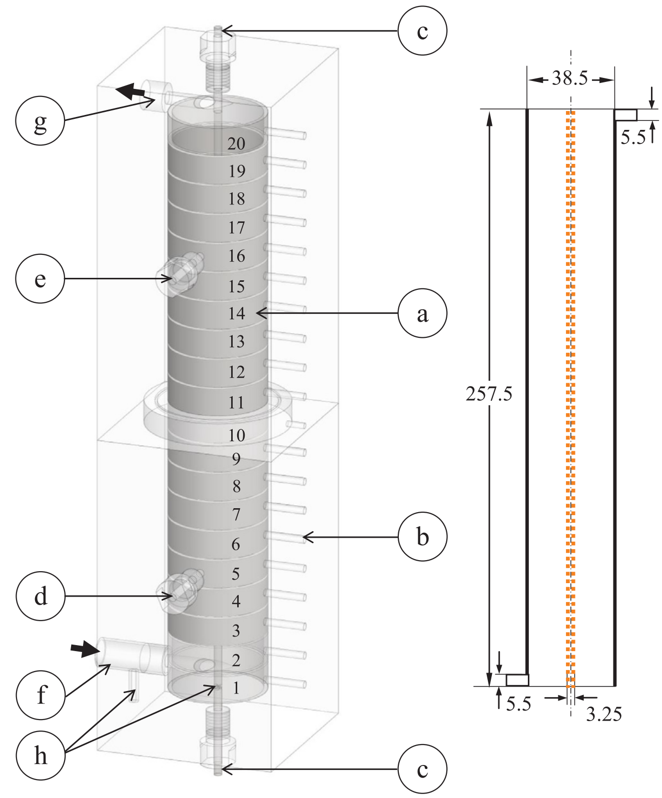

Fig. 1. Schematic representation of the decaying swirling flow electrochemical reactor. (a) Segmented cathode; (b) electrical connections to calibrated resistors; (c) electrical connection to anode; (d) Haber-Luggin capillary for cathodic potential control; (e) second Haber-Luggin capillary for measurement of the cathodic potential; (f) tangential electrolyte inlet; (g) tangential electrolyte outlet; (h) nitrogen feeder for biphasic system. The numbers indicate the position of the segments and the arrows the flow of the electrolyte. Right hand side: thick full line: cathode; thick dotted line: anode. Internal dimensions of the reactor in mm.

The electrical contact to the external electrode was made by a screw through the acrylic wall pressing the backside of each segment. Calibrated resistors, 0.025 Ω resistance, join the screws of each segment and the cathodic current feeder, which was electrically connected at both ends. By measuring the ohmic drop in the resistors, it was possible to determine the axial current distribution and to calculate the local mass-transfer coefficient at each segment. The effect of the calibrated resistors on the current distribution can be neglected due to the small value of their ohmic drop, lower than 5 mV, in comparison with the other terms of the voltage balance in the reactor. The data acquisition was performed using a computer controlled home-made analogue multiplexer and five independent data sets were obtained for each