

Mass Transport in the Rectangular Channel of a Filter-Press Electrolyzer (the FM01-LC Reactor)

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The FM01-LC is a laboratory-scale (projected area 64 cm²) electrochemical cell, based on the larger (2100 cm²) FM21-SP electrolyzer (INEOS Chlor-Chemicals), the latter having originally been designed for chlor-alkali processing.¹ Both of these electrochemical cells are designed with a plate-and-frame filter-press configuration; electrodes, spacers, ion-exchange membranes and gaskets are compressed between two electrically insulating end plates. The available electrode area in the FM01-LC reactor can be readily increased by building up a series of standard sized electrode plates, or by the use of additional stacks. The reactor is well suited to laboratory feasibility studies in subject areas, such as electrosynthesis, environmental treatment and redox flow cells, together with investigations of the effect of process variables on electrochemical reactions.^{1, 2}

A number of articles on the characterization of FM01-LC reactors have been published.¹⁻⁷ Brown et al.⁴ have examined the current distribution within the reactor for the reduction of cupric ions to copper metal, showing that plastic mesh turbulence promoters had the effect of smoothing out the current distribution within the cell and reducing entry effects. The same authors studied the space-averaged mass transport of the FM01-LC using the reduction of ferricyanide ion at nickel electrodes.³ They examined the relationship between the mass-

transport coefficient and the mean linear flow velocity (with and without turbulence promoters). Increasing the velocity of the electrolyte increased the rate of mass transport at the expense of a higher pumping cost and a lower reactant conversion per pass. The space averaged mass-transport coefficient improved by a factor of 1.7 to 3.8. The pressure drop across the cell was small (and dominated by the restrictions imposed by the manifolds at the entrance and outlet of the cell). A study using three-dimensional (3-D) electrodes occupying the entire electrolyte channel of the FM01-LC cell has been published by the same authors,⁵ the mass transport characteristics of different 3-D electrode materials were compared using the reduction of ferricyanide ion as a model reaction. The authors concluded that the FM01-LC can be easily modified to use 3-D electrodes, which allows the overall reaction rate to be enhanced by a factor of up to 100 times. The enhanced surface area was used to oxidize alcohols and, hence, synthesize carboxylic acids.

Trinidad and Walsh⁶ examined the effect of 3-D electrodes and turbulence promoters on the fluid-flow distribution within the FM01-LC. Deviations from the ideal plug-flow model were quantified for different reactor models by plotting the Peclet number vs. electrolyte velocity. Turbulence promoters increased the mass transport leading to a more uniform current density, with reduced entrance effects; the plug flow model was found to be applicable when suitable turbulence promoters are used. An article by the same authors⁷ described the oxidation of cerous ions using the FM01-LC in a divided mode. The authors demonstrated the feasibility of the process and characterized reactor performance in terms of reactant conversion. A study of flow visualization and residence time distribution (RTD) of the FM01 cell in an empty channel, and with foam electrodes as a

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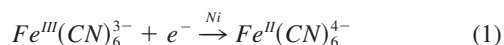
turbulence promoters showed a significant increase of the Peclet number compared to an empty channel.⁸

Other applications of the FM01-LC reactor include: the condensation of carboxylic acids (Kolbe synthesis), the oxidation of dinitrogen tetroxide to dinitrogen pentoxide,⁹ the breaking of the nitric acid azeotrope,¹⁰ the electrosynthesis of coumestan and catecholamine derivatives, using carbon electrodes,¹¹ and the generation of hydrogen from a 10 wt % aqueous sulfuric acid solution using a pulsed direct current.¹²

In this article, the effect on mass transport and pressure drop has been examined when the mean linear flow velocity of the electrolyte was taken to appreciably higher values than in the previous works. The opportunity was also taken to use a sodium carbonate background electrolyte, which provides a much more stable background electrolyte,¹³ than the previously used⁴ potassium hydroxide.

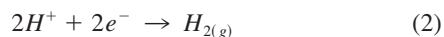
Theory

Mass transport characterization of the FM01-LC reactor can be considered using the a model reaction, namely, the reduction of ferricyanide ion to ferrocyanide ion, at a nickel cathode, by measuring the (convective-diffusion controlled) limiting current over a range of flow rates of electrolyte. The reaction



has been widely used to characterize mass transport in electrochemical reactors.^{2,3,11,14,15} Under complete mass transport control, the limiting current value represents a maximum flux of material toward the electrode surface at a particular flow rate. Beyond the limiting current region, plateau region, the potential is sufficiently negative for secondary reactions (typically hydrogen evolution) to occur, and the current then continues to increase.

The most common secondary reaction, which would be expected to occur at the nickel cathode, is the evolution of hydrogen gas



Under full mass transport conditions, that is, in the limiting current plateau region, the current is related to the global mass-transport coefficient k_m

$$k_m = \frac{I_l}{AzFc} \quad (3)$$

Here, I_l is the limiting current for reduction of ferricyanide ions, A is the electrode area, z is the number of electrons transferred, F is the Faraday constant ($96\,485\text{ C mol}^{-1}$), and c is the concentration of the electroactive species in the bulk of the electrolyte.

Experimental Details

The undivided mode with a single combined anolyte and catholyte compartment and the electrolyte flow circuit for the FM01-LC cell is shown in Figure 1a, together with the electrical circuit used. The electrolyte was contained in a 1.5 dm^3

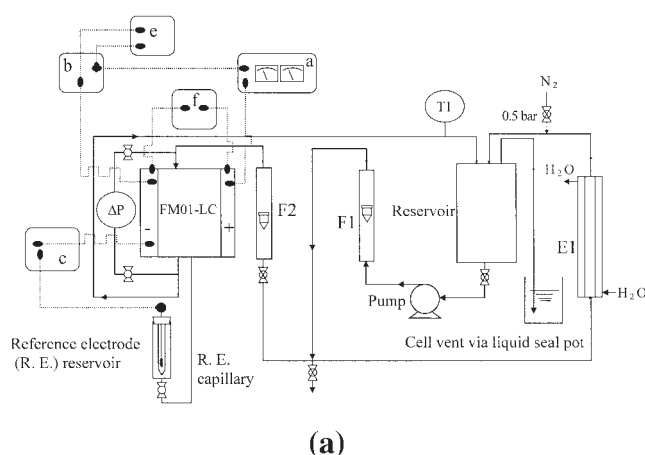


Figure 1a. Electrical and flow circuits for the measurement of mass transport at the cathode of an FM01-LC electrochemical reactor.

(a) dc power supply unit; (b) standard shunt (200 mA/200 mV); (c) digital multimeter (cathode potential measurement); (d) saturated calomel electrode (SCE); (e) digital multimeter (current measurement), and (f) digital multimeter (cell voltage measurement).

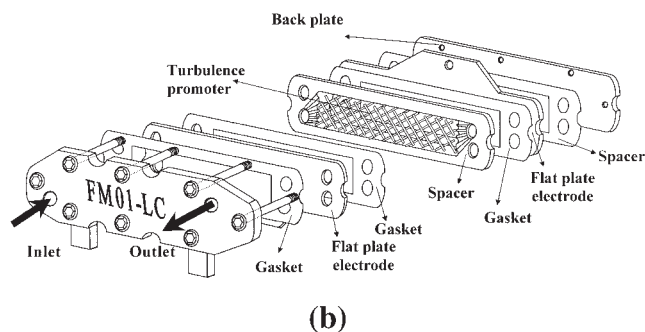


Figure 1b. Exploded view of the FM01-LC laboratory cell electrolyzer in the undivided mode including the plastic mesh turbulence promoter type D in reference [16].

glass reservoir, sealed from the surroundings and vented by a tube through a liquid seal pot. A magnetically coupled, centrifugal pump was used to circulate electrolyte through the cell. In previous work by Brown et al.,^{3-5, 16} experiments were conducted at linear electrolyte velocities in the range 2.4 to 9.7 cm s^{-1} . In this article, an appreciably higher maximum velocity was used, the range being 5.2 to 22.4 cm s^{-1} .

The electrolyte circuit was constructed from PTFE tubing and fittings. The cell was uniformly compressed to a torque at 25 Nm , giving a measured interelectrode gap of 0.55 cm . An exploded view of the cell that includes the turbulence promoter used within the cell channel is shown in Figure 1b, the turbulence promoter is a plastic routed mesh, promoter type D in reference.¹⁶ The promoter was 0.55 thick and a single sheet of the PTFE polymer filled the channel completely, details of the characteristic of the turbulence promoter are given in Table 1. A notch of the plastic was removed to allow the Luggin capillary to protrude into the compartment. Flow rates were measured using a variable area, glass rotameter.

The temperature of the electrolyte was monitored to ensure

Table 1. Nickel Electrode Dimensions, Experimental Details of the FM01-LC Reactor, Electrolyte Properties, and Temperature

Electrode length, L	16 cm
Electrode height, B	4 cm
Electrode spacing, S	0.55 cm
Electrode area, A	64 cm ²
Hydraulic (equivalent) diameter, $d_e = 2BS/(B + S)$	0.97 cm
Faraday constant, F	96,485 C mol ⁻¹
Number of electrons transferred per ion, z	1
Density of electrolyte, ρ	1.0985 g cm ⁻³
Kinematic viscosity of electrolyte, μ	0.00956 cm ² s ⁻¹
Diffusion coefficient of Fe(CN) ₆ ³⁻ , D	6.4×10^{-6} cm ² s ⁻¹
Schmidt number of electrolyte, $Sc = \nu/D$	1494
Electrolyte composition	1 mM $K_3Fe(CN)_6$ + 10 mM $K_4Fe(CN)_6$ + 1 M Na_2CO_3
Mean fluid velocity, v	5.2 to 22.4 cm s ⁻¹
Temperature	302 K
Turbulence promoter	Plastic mesh type D *CD and **LD = 11 mm. Overall voidage 0.83

* CD = internal dimension of shorter mesh diagonal.

**LD = internal dimension of longer mesh diagonal.

Overall voidage is the ratio of free space in the channel to overall channel volume.

that steady-state conditions had been reached. A controlled potential difference was applied across the cell using a power supply (Thurlby Thandar Instruments TSX1820 Precision DC PSU). Current was via monitoring the potential difference across a standard shunt (200 mA/200 mV) using a digital voltmeter (dvm). The cathode potential was measured between the nickel electrode and the saturated calomel electrode (SCE) using a high impedance dvm, and the cell potential was measured between the anode and the cathode by third dvm.

The concentration of the electroactive species (ferricyanide ion) was 1 mM (as the potassium salt). In order to ensure that the rate of the anode reaction (oxidation of ferrocyanide to ferricyanide) never becomes the limiting reaction, potassium ferrocyanide was present in excess. It has been suggested¹⁷ that a ferrocyanide/ferricyanide ratio of 5:1 is sufficient to ensure that ferrocyanide ion was present at a concentration of 10 mM. The background electrolyte was 1M sodium carbonate. According to Taama et al.,¹³ who studied the effect of supporting electrolyte on ferricyanide reduction at a rotating disc electrode, potassium carbonate provides much more stable limiting current vs. time behavior than potassium hydroxide, despite the extensive use of KOH in previous work^{2, 3, 15, 18}. Two liters of electrolyte were freshly prepared for each experiment to fill the reservoir and allow for makeup, if required, during the experiment. The electrolyte solution was prepared using freshly deionized water. In order to minimize ultraviolet degradation of the electrolyte during the course of the experiments, the electrolyte reservoir and reference electrode flask were shrouded in aluminum foil. The electrolyte was drained from the system at the end of each day of studies and a fresh batch of electrolyte was prepared at the start of each experiment.

The electrodes were pretreated to provide a uniform, clean surface for mass transport before installing them in the cell. The nickel electrode and its contacts were polished with low grit grade sand paper and rinsed with deionized water. A planar niobium electrode plated with platinum was used as a counter electrode. A fast stream of nitrogen was used to deoxygenate the electrolyte during 20 min before each experiment. Oxygen remained excluded as the circuit was sealed from the atmosphere by a liquid seal pot containing the working electrolyte (Figure 1a). Current vs. electrode potential data were obtained

over a range of flow rates in an empty channel, and with the turbulence promoter mesh present.

The open-circuit potential for the nickel electrode in the ferri/ferrocyanide system with a 1M KOH background electrolyte is reported as being +0.22 V vs. SCE at 25°C.³ In previous work,¹⁶ a potentiostat was used to scan the cathode potential from the open-circuit potential to a value of -1.1 V vs. SCE, at a rate of 3 mV s⁻¹. Hydrogen evolution was reported as occurring between 0.9 V and 1.0 V vs. SCE. In the experiments reported in the literature, current-potential curves were recorded on an x-y chart recorder. In this experiment, the cell potential was increased manually in 50 mV steps up to -1.333 V, and the corresponding current and cathode potential (vs. SCE) was recorded. The cathode potential ranged from the open circuit potential (which was in the range +0.190 and +0.160 V vs. SCE, depending on the electrolyte flow rate) to approximately -1.1 V vs. SCE. In this article, potentials ranged from -0.19 V vs. SCE to +1.1 V vs. SCE.

Once a family of steady-state current vs. time curves had been recorded, a constant potential value was selected on the plateau region, and the current is measured at a range of flow rates. A graph of current vs. electrolyte velocity (or Reynolds number) could then be drawn to show the relationship between linear electrolyte velocity and limiting current then utilized to find the mass-transfer coefficients over the range of fluid velocities.

All results were obtained at steady state; voltage readings were allowed to stabilize before being recorded. The temperature in all the experiments was 29±2 °C. The physical and transport properties of the electrolyte used to calculate dimensionless groups and mass-transport coefficients are given in Table 1. The density of the electrolyte was measured at 29°C, with the aid of a hot water bath, using a density meter (Anton Paar DMA 35).

Pressure drops across the cell, with and without a mesh turbulence promoter, were measured at different mean linear flow velocities. For the pressure drop measurements, the maximum linear velocity was restricted by the size of the manometer available. A 1 m tall, vertical glass, u-tube, water-filled manometer was employed, using water as the fluid. The manometer was connected by silicone tubing to tees located as close as possible to the inlet and outlet of the cell. Several measurements of pressure drop across the cell were taken for

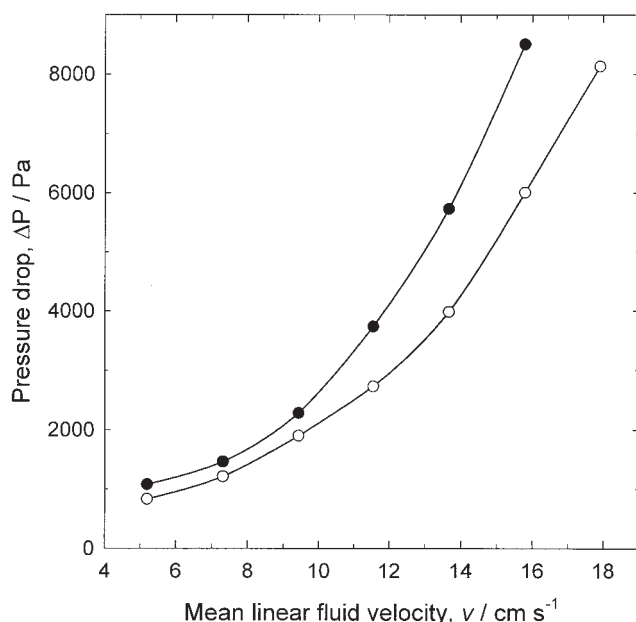


Figure 2. Pressure drop across the FM01-LC with varying linear fluid velocity for turbulence promoted (●) and empty (○) channel.

each mean linear flow rate, and the averaged used in subsequent analysis. The same procedure was then applied when the mesh turbulence promoter was incorporated into the cell.

Results and Discussion

Pressure drop

Figure 2 shows the pressure drop vs. mean linear fluid velocity; as predicted, the pressure drop across the cell increased with linear fluid velocity, and was higher for the promoted channel than for the empty channel, due to the increased turbulence. The increase in pressure drop resulting from the installation of a mesh turbulence promoter may also be charted against linear velocity, by subtracting the pressure drop for the empty channel from the pressure drop for the promoted channel at each velocity. This relationship shows an exponential increase in pressure drop with increasing electrolyte velocity. The penalty paid in terms of increased pressure drop for installing a mesh turbulence promoter in the cell increases dramatically.

In previous studies, Brown et al.^{4, 16} plotted the logarithm of the pressure drop against the logarithm of Reynolds number, in order to fit the experimental data to power law relationship of the form

$$\Delta P = a Re^b \quad (4)$$

where a and b are 0.59×10^2 and 1.87, respectively for an empty channel in this work. The Reynolds number at each linear fluid velocity is determined

$$Re = \frac{vd_e}{\nu} \quad (5)$$

where v is the mean linear fluid velocity of the fluid through the cell, d_e is the equivalent (hydraulic) diameter of the flow channel, and ν is the kinematic viscosity; the latter two terms are given in Table 1. The power relationship in Eq. 4 with and without turbulence promoter was $\Delta P = 0.63 \times 10^{-2} Re^{1.89}$, and $\Delta P = 0.59 \times 10^{-2} Re^{1.87}$, respectively. For both the empty and the turbulence promoted channel, the value of the constant “ a ” is smaller than the literature values. This is counterbalanced by the index “ b ” being larger for the derived results than in the literature.^{3, 4}

Mass transport

Mass transport was characterized by measuring the limiting current over a range of electrolyte velocities, both with and without the presence of mesh turbulence promoters, by the method described in the previous section. In the absence of turbulence promoter at a mean linear flow rate of 9.4 cm s^{-1} the limiting current plateau appeared in the same region when both cell and cathode potentials were monitored separately. Therefore, either potential may be measured against current in order to determine the limiting current values for the reduction of ferricyanide ion in a sodium carbonate supporting electrolyte.

The effect of increasing the mean linear flow velocity on the limiting current is shown in Figure 3 for the empty channel. The figure shows that, in all cases, increasing the electrolyte velocity results in an enhanced limiting current. A similar trend was observed when the turbulence promoter was installed in the cell but the currents were approximately 25–30% higher.

A useful figure of merit is the mass transport enhancement factor, γ , which can be defined¹⁵ as

$$\gamma = \frac{k_{L,\text{promoted}}}{k_{L,\text{unpromoted}}} = \frac{I_{L,\text{promoted}}}{I_{L,\text{unpromoted}}} \quad (6)$$

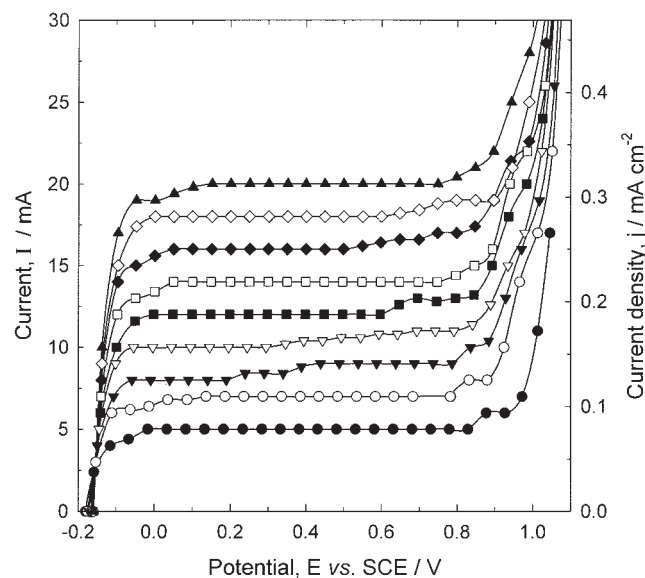


Figure 3. The effect of increasing mean linear fluid velocity on the current vs. cell potential curves in the absence of a turbulence promoter (empty channel).

Mean linear flow velocity: ● 5.2, ○ 7.3, ▼ 9.4, ▽ 11.6, ■ 13.7, □ 15.8, ◆ 17.9, ◇ 20.0, ▲ 22.1 cm s^{-1} .

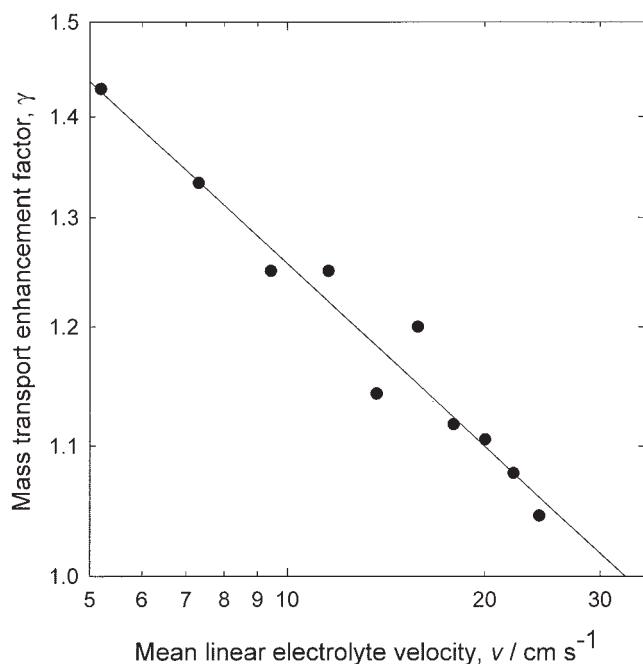


Figure 4. Log-log plot of mass transport enhancement factor (γ) vs. mean linear electrolyte velocity, extrapolated to the intercept with $\gamma = 1$ (no enhancement).

The semilog plot of γ vs. electrolyte velocity is given in Figure 4. The mass transport enhancement factor declines with increasing velocity toward $\gamma = 1$, that is, no enhancement due to the presence of a turbulence promoter. The relationship between the mass transport enhancement, and the mean linear electrolyte velocity v is: $\gamma = 1.96 v^{-0.192}$, with a linear correlation coefficient of 0.95. The enhancement factor is 1 when the mean linear velocity is 33 cm s^{-1} . This means that, at higher electrolyte velocity, no benefit would be gained in terms of the mass transport from the installation of a mesh turbulence promoter; the studies by Brown et al.⁵ support this conclusion.

For industrial-scale applications, the actual upper electrolyte velocity for which a mesh turbulence promoter would be beneficial would be dependent on an economic trade-off. The enhanced mass transport rates provided by the use of plastic mesh turbulence promoters must be balanced against the increased pumping costs associated with a higher-pressure drop across the cell.

The fluid velocity range used by Brown et al.^{3-5, 16} was lower than in this work, but when velocities are similar (5.2 to 9.4 cm s^{-1}), it was found that in each case the literature values of mass-transport coefficient were higher than the experimental values reported here. The slope of the curve of mass-transport coefficient vs. electrolyte velocity was also twice as steep for the literature values. Using the properties of the electrolyte reported in Table 1, dimensionless groups were calculated for the mass transport data, in order to fit them to a correlation of the form

$$\text{Sh} = a \text{Re}^b \text{Sc}^{0.33} \quad (7)$$

This correlation is widely used in the literature^{3, 13, 16, 18} for representing mass transport in electrochemical reactors. The

Reynolds number was given in Eq. 5, and the Sherwood number is defined by

$$\text{Sh} = \frac{k_m d_e}{D} \quad (8)$$

That is, channel hydraulic dia. d_e is 0.97 cm , the Schmidt number is given in Table 1. Arranging Eq. 7 in its logarithmic form

$$\log \text{Sh} = b \log \text{Re} + \log a + 0.33 \log \text{Sc} \quad (9)$$

The linear correlation from the log-log plot of Sh vs. Re shown in Figure 5 for both the empty channel and the turbulence promoted channel allows to determine the constants a and b and \cdot . For the empty channel, “ a ” was found from the inverse logarithm of the y-axis intercept after subtracting $0.33 \log(\text{Sc})$, and equals 0.18 . The index “ b ” is the gradient of the trend line, and equals 0.73 . Similarly, for the channel with the mesh turbulence promoter, “ a ” was found to be 0.71 and “ b ” was found to be 0.55 . These values are summarized in Table 2, where they are compared with literature values for the FM01-LC, and ideal fully developed laminar and turbulent flow, are plotted in Figure 5, as well.

The experimentally determined Sherwood - Reynolds correlations were broadly comparable to the literature correlations for the FM01-LC reactor. Sherwood number values were higher with a mesh turbulence promoter installed than without, and all were considerably higher than the theoretical “fully-developed” laminar and turbulent flow. Over the range of Reynolds numbers for which

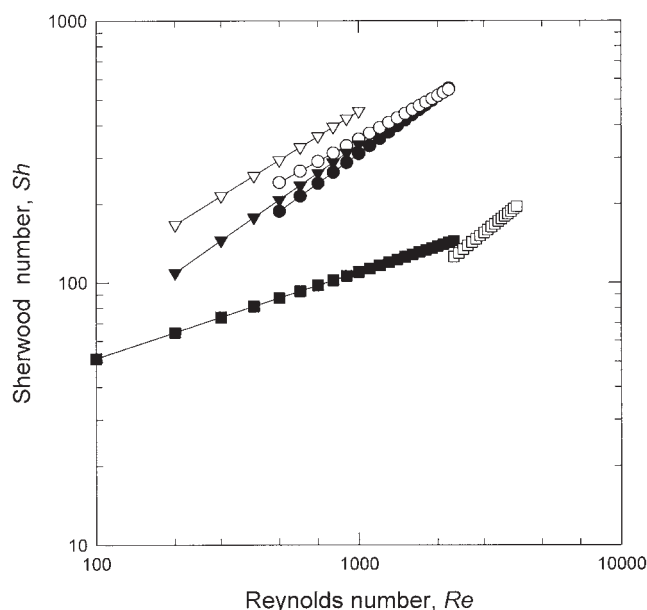


Figure 5. Log of Sherwood vs. log of Reynolds numbers for experimentally determined (and literature) constants of the FM01-LC reactor, together with theoretical laminar and turbulent flow constants.

● This work without turbulence promoter (empty channel), ○ this work with turbulence promoter, ▼ without turbulence promoter (empty channel),⁵ ▽ with turbulence promoter type D,⁵ ■ fully developed laminar flow,⁵ □ fully developed turbulent flow.⁵

Table 2. Experimental and Literature Values of the Constants a , b , and c in the Sherwood Correlation ($Sh = a Re^b Sc^c$) for the FM01-LC Reactor, Together with Literature Values for Idealised, Fully Developed Laminar and Turbulent Flow

Source of Dimensionless Mass Transport Correlation	a	b	c	Re
Experimental FM01-LC with no turbulence promoter	0.18	0.73	0.33	500–2200
Experimental FM01-LC with mesh turbulence promoter	0.71	0.55	0.33	500–2200
Literature values [19]. FM01-LC with no turbulence promoter	0.24	0.7	0.33	200–1000
Literature values [19]. FM01-LC with type D turbulence promoter	0.56	0.62	0.33	200–1000
Ideal values [19]. Fully developed laminar flow.	2.54	0.33	0.33	<2300
Ideal values [19]. Fully developed turbulent flow.	0.023	0.8	0.33	>2300

experiments were conducted ($Re = 500 - 2200$), the flow would be expected to be laminar. Brown et al.,¹⁶ however, suggests that a nonlaminar flow regime should in fact be expected as the cell has internal manifolds, and no calming section between the inlet and electrode face, and this causes the steeper slopes.

Conclusions

Measurements of mass transport and pressure drop on the FM01-LC cell (both with turbulence promoter and in an empty rectangular channel) have been investigated at higher mean fluid velocity of the electrolyte than in previous work reported in the literature. The average mass-transport coefficient was calculated using the limiting current technique, using the reduction of 1mM ferricyanide ion in 1M sodium carbonate at 29°C.

The pressure drop with and without turbulence promoter appears to increase exponentially. Furthermore, the increase in pressure drop resulting from the installation of a turbulence promoter increased pressure drop dramatically with the mean linear flow rate.

Mass-transport coefficients were found to improve through the presence of a mesh turbulence promoter. The mass transport enhancement factor due to the mesh turbulence promoter γ was found to decrease with increasing linear velocity. A value of 33 cm s^{-1} for the mean fluid velocity was the limit beyond which there is no advantage in increasing velocity as the mass transport enhancement factor reaches a value of 1.

The mass transport characteristics have been shown to fit the following Sherwood correlation. For an empty channel

$$Sh = 0.18 Re^{0.74} Sc^{0.33} \quad (10)$$

For the turbulence promoted channel the mass transport correlations was

$$Sh = 0.71 Re^{0.55} Sc^{0.33} \quad (11)$$

These correlations differ by a maximum of 20% compared with the correlations reported in the literature for a similar electrochemical system (see Table 1⁴, 16, 18) despite the fact that in this work a substantially higher mean linear velocity has been used (5.2 to 22.1 cm s^{-1}). This difference can be caused by several factors including (a) changes in the area of the electrode, (b) differences in the preparation of the electrolyte, and (c) the use of sodium carbonate as a supporting electrolyte.

Acknowledgments

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