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## Short Communication

# Mass transport evolution in microfluidic thin film electrochemical reactors: New correlations from millimetric to submillimetric interelectrode distances

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## ABSTRACT

Correlation of the mass transfer characteristics of microfluidic parallel-plate electrochemical reactors is proposed for the first time. Firstly, the variation in the mass transfer coefficient ( $k_m$ ) ( $1.61\text{--}3.94 \times 10^{-5} \text{ m s}^{-1}$ ) over a wide range of interelectrode distances ( $d_{\text{elec}}$ ) from millimetric (3 mm) to micrometric values (100  $\mu\text{m}$ ) is reported. Secondly, a drastic slope change in the curve makes it possible to identify the onset of microfluidic behavior in a quantitative way for the first time, i.e. below a 1000  $\mu\text{m}$  gap. Thirdly, a mathematical model is proposed which predicts  $k_m$  for any  $d_{\text{elec}}$  of interest. Fourthly, under laminar flow ( $7 < \text{Reynolds } (Re) < 623$ ) and for temperatures in the range of 10–50 °C ( $532 < \text{Schmidt } (Sc) < 3315$ ), new Sherwood ( $Sh$ ) correlations are obtained for both microfluidic and millimetric configurations. It is thus feasible to extrapolate  $k_m$  for microfluidic electrochemical reactor scale-up.

## 1. Introduction

Microscale electrochemical reactors provide an interesting approach in various disciplines, such as electrosynthesis [1–5], electrodeposition [6,7], sensor development [8,9] and wastewater treatment [10–15]. Versatility, a small footprint, suitability for reactions with rapid kinetics, easy thermoregulation, independence of supporting electrolyte, mass transfer intensification and huge energy savings are among the benefits to be gained from microfluidic flow reactor technology [16–18]. Measurement of mass transfer in the submillimetric range has been studied since the early 1970s, with the development of flow rectangular channel, tubular and granular bed electrochemical reactors [19–27]. Estimating the average mass transfer coefficient ( $k_m$ ) is particularly crucial in the sizing of electrochemical reactors and for scale-up studies [28]. In the case of millimetric scale reactors, the correlation of mass transfer with different geometries with or without turbulence promoters [28–33], with different electrode morphologies [34,35] as well as innovative electrolyte inlets [36,37] and agitation systems [38,39] has been widely investigated in the literature. However, more data are required on mass transfer evolution and correlation in microfluidic parallel-plate reactors at a larger scale [40]. This could be due to problems in carrying out experiments under different hydrodynamic

conditions and temperatures as well as to the difficulty of configuring 3-electrode setups with a narrow interelectrode gap [38,39]. In this paper, mass transfer evolution over a wide range of interelectrode distances ( $d_{\text{elec}}$ ) from millimetric (3 mm) down to submillimetric (100  $\mu\text{m}$ ) values is reported for the first time, using a flow-by parallel-plate reactor with a large effective surface area. This reactor design is suitable for scaled-up industrial applications. A model has been proposed which evaluates  $k_m$  as a function of  $d_{\text{elec}}$ . Finally, a mass transfer correlation involving dimensionless numbers in microfluidic and millimetric systems is proposed.

## 2. Materials and methods

## 2.1. Electrochemical system

The electrolyte (0.5 L) was stored in a double-jacket stainless steel reservoir connected to a thermostatic bath (Bioblock Scientific, Poly-Science, Niles, IL, USA) which regulates the working temperature between 10 and 50 °C in this series of experiments. A peristaltic pump (Masterflex, Cole-Parmer, Vernon Hills, IL, USA) was used to circulate the electrolyte at the desired flow rate in batch recirculation mode. The electrochemical setup featured a parallel-plate reactor (a schematic

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illustration is given in Fig. S1 in the Supplementary information). Both working and counter electrodes were planar rectangles, facing one another. 316L stainless steel (SS) (Gantois Industries, Saint-Dié-des-Vosges, France), 5 mm in thickness, was used as the working electrode. Boron-doped diamond (BDD) (DiaCCon, Fürth, Germany), 2 mm in thickness, coated on both sides with a niobium substrate 12  $\mu\text{m}$  thick, served as the counter electrode. Their width ( $W$ ) and length ( $L$ ) were 5 and 10 cm respectively, giving an effective surface area of 50  $\text{cm}^2$ . Spacers made of polytetrafluoroethylene (PTFE) with different thicknesses (100, 250, 500, 1000, 1500, 2000 and 3000  $\mu\text{m}$ ) from Bohlender, Grünsfeld, Germany, were used to separate the working and counter electrodes. These therefore defined the values of  $d_{\text{elec}}$  investigated in this work, from the micrometric scale progressively into a typical millimetric configuration. Silver-saturated silver chloride ( $\text{Ag-AgCl/Cl}^-$ ) was used as the reference electrode. This was inserted in the lower part of the cell, near the electrolyte inlet stream. The electrolyte was introduced by forced convection from the bottom of the cell, flowing upward against the gravitational force. The positioning of the reference electrode (lower part of the cell, Fig. S1) takes advantage of the gravitational force to ensure stable measurement conditions by avoiding both engorgement in the case of unstable electrolyte flow and perturbation due to gas evolution at very low applied potentials. Furthermore, the  $\text{Ag-AgCl}$  reference electrode was positioned very close to the working electrode (500  $\pm$  100  $\mu\text{m}$ ) in order to avoid ohmic loss in solution [41]. The BDD electrode features a wide potential window of water stability with the  $\text{O}_2$  evolution reaction (OER) occurring at 2.5–2.7 V/SHE [42,43]. At this high potential OER, negligible interference from  $\text{O}_2$  evolved on the counter electrode may be expected.

## 2.2. Mass transfer characterization

The mass transfer in the electrochemical cells was studied using the widely used limiting-current technique described elsewhere [41,44]. 0.5 L of electrolytic solution containing 0.05 M  $\text{K}_3\text{Fe}(\text{CN})_6$ , 0.10 M  $\text{K}_4\text{Fe}(\text{CN})_6$  and 0.50 M  $\text{Na}_2\text{CO}_3$  as supporting electrolyte was employed. A high concentration of supporting electrolyte was used to avoid the electro-migration effect [41,45]. Using this technique, the reduction of ferricyanide ( $\text{Fe}(\text{CN})_6^{3-}$ ) achieved its maximum rate (limiting current plateau) so that the reduction was purely controlled by the mass transport regime. The physical properties of the electrolyte at the different temperatures under investigation [29,30,46] are given in Table S1 in the Supplementary information. Linear scan voltammetry (LSV) was carried out using a potentiostat (Ametek, Massy, France) equipped with a PMC-1000 channel that can perform both DC and AC experiments up to  $\pm 10$  V and  $\pm 2$  A, respectively. LSV was used to study the cathodic potential activity of the ferri-ferrocyanide electrolyte over a range of potentials from 0.2 to  $-1.6$  V versus the  $\text{Ag/AgCl}$  reference electrode at a scan rate of 2.5  $\text{mV s}^{-1}$ . Within the chosen range of flow rates, the scan rate was sufficiently slow to attain a steady state [47]. To check the influence of any dissolved  $\text{O}_2$  in the electrolyte, preliminary experiments were conducted at both extremes (i.e. 100 and 3000  $\mu\text{m}$ ) as well as using a 500  $\mu\text{m}$  interelectrode gap after the electrolyte had been purged of dissolved  $\text{O}_2$ . The electrolyte in the reservoir was aerated with  $\text{N}_2$  gas for 30 min before the LSV to eliminate dissolved  $\text{O}_2$ . The results indicated that the limiting current plateau (just before the reduction of the solvent) was not influenced by the presence of dissolved  $\text{O}_2$  (data not shown). The resulting limiting current values ( $I_{\text{lim}}$  in A) for different  $d_{\text{elec}}$  under the chosen operating conditions were related to the average  $k_m$  ( $\text{m s}^{-1}$ ) via Eq. (1) [44]:

$$k_m = \frac{I_{\text{lim}}}{nFSC_{\text{sol}}} \quad (1)$$

where  $n$  is the number of electrons involved in the reduction reaction of ferri- to ferrocyanide (i.e. 1),  $F$  is Faraday's constant ( $96485 \text{ C mol}^{-1}$ ),  $S$  is the effective surface area of the working electrode ( $\text{m}^2$ ) and  $C_{\text{sol}}$  is the

molar concentration of ferricyanide in the bulk solution ( $\text{mol m}^{-3}$ ).

The flow rate varied from 10 to 600  $\text{mL min}^{-1}$ , when  $d_{\text{elec}}$  was increased from 100 to 3000  $\mu\text{m}$ . Across all investigated values of  $d_{\text{elec}}$  and for relevant comparison, the electrolyte flow was kept within the laminar regime with Reynolds ( $Re$ ) values in the range 7 to 623. Owing to the entrance manifolds of the tube-type entry of the electrochemical cell, under similar hydrodynamic regimes, the electrolyte recirculating zone in the reactor was considered minimal in all investigated configurations [36,48,49].

## 3. Results and discussion

### 3.1. Mass transfer behavior over a wide range of interelectrode distances

Average  $k_m$  values over the wide range of  $d_{\text{elec}}$ , obtained from the plateau of  $I_{\text{lim}}$  identified on voltammograms for the different values of  $d_{\text{elec}}$  (Fig. S2), are shown in Fig. 1. Interestingly, two distinct behaviors of  $k_m$  were observed as a first approach. The increase in  $k_m$  in the millimetric range from 3000  $\mu\text{m}$  down to 1000  $\mu\text{m}$  seemed linear (slope:  $-10^{-3} \text{ s}^{-1}$ , coefficient of determination  $R^2 = 0.91$ ). For values lower than 1000  $\mu\text{m}$ ,  $k_m$  increased drastically as  $d_{\text{elec}}$  decreased into the micrometric range. This trend suggests that the mass transfer was massively enhanced while transitioning from a millimetric to a micrometric gap, which supports the interest in microfluidic reactors for transfer intensification [14,16,50–55]. The range of values of  $k_m$  determined under both millimetric and micrometric configurations in this work are in close agreement with those reported in the literature (Table S2 in the Supplementary information). In the millimetric range,  $k_m$  values of  $1.61 \times 10^{-5} \text{ m s}^{-1}$  and  $1.82 \times 10^{-5} \text{ m s}^{-1}$  were obtained for 3000  $\mu\text{m}$  and 1000  $\mu\text{m}$ , respectively, compared with reported values of  $k_m$  between 0.30 and  $1.20 \times 10^{-5} \text{ m s}^{-1}$  for 5000  $\mu\text{m}$  [56] and  $1.60 \times 10^{-5} \text{ m s}^{-1}$  for 1000  $\mu\text{m}$  [28].

The Levich equation (Eq. (2)) has been widely used in the literature to relate  $I_{\text{lim}}$  to electrolyte volume flow rate in thin rectangular channel electrode reactors [19,57,58].

$$I_{\text{lim}} = 0.925nFWC_{\text{sol}}(D_L X_c)^{2/3} \left( \frac{\dot{V}_F}{h^2 d} \right)^{1/3} \quad (2)$$

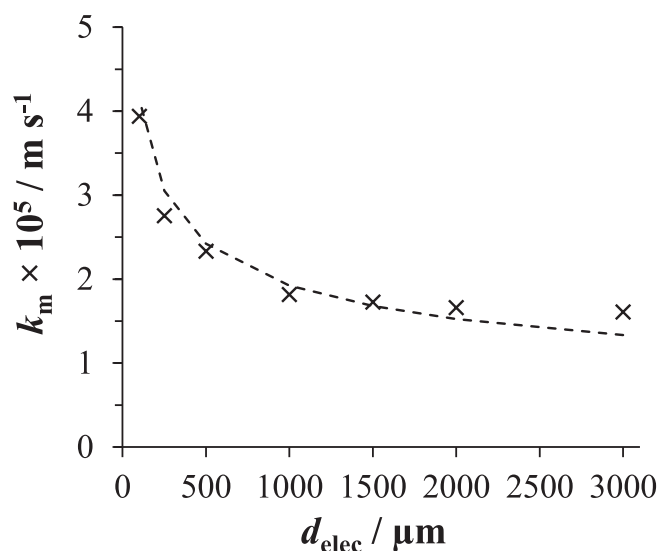


Fig. 1. Variation of experimental and theoretical average values of  $k_m$  as a function of  $d_{\text{elec}}$ . Operating conditions: Flow rate: 10–600  $\text{mL min}^{-1}$ , temperature: 25  $^{\circ}\text{C}$ , identical cross-sectional electrolyte velocity ( $u_1$ ): 4  $\text{m min}^{-1}$  and electrolyte residence time ( $\tau$ ): 0.025 min (see Text S1 in the Supplementary Information).

where

$$\dot{V}_F = u_L W d_{\text{elec}} \quad (3)$$

$$h = \frac{d_{\text{elec}}}{2} \quad (4)$$

Here  $W$  is the electrode width (in m),  $D_L$  is the diffusion coefficient (in  $\text{m}^2 \text{s}^{-1}$ ),  $x_c$  is the electrode length (equivalent to  $L$ , in m),  $\dot{V}_F$  is the volumetric flow rate (in  $\text{m}^3 \text{s}^{-1}$ ),  $h$  is half the cell depth (in m),  $d$  is the channel width (equivalent to  $W$  in this work, in m) and  $u_L$  is the cross-sectional velocity (in  $\text{m s}^{-1}$ , defined as Eq. S1 in the [Supplementary Information](#)). Equation (2) is ideally applicable under conditions where  $h \ll d$ , there is a sufficiently large electrode area, the electrolyte flow rate is not too low and the profile of electrolyte flow velocity in the channel is parabolic [19]. Relating Eq. (1) and Eq. (2) yields an expression of average  $k_m$  as a function of  $d_{\text{elec}}$  (Eq. (5)):

$$k_m = \frac{2.94 D_L^{2/3} u_L^{1/3} W L^{2/3}}{S d_{\text{elec}}^{1/3}} \quad (5)$$

A slight adjustment of the coefficient in Eq. (2) was made to match the experimental data. This deviation could be due to the fact that direct application of Eq. (2) is not ideal under conditions where the electrode width and channel width are identical ( $W = d$ ), in which an edge effect could not be neglected [19,40]. The plot of experimental and theoretical  $k_m$  versus  $d_{\text{elec}}$  is given in Fig. 1. The theoretical curve matches its experimental counterpart well, with a deviation of 0.000187 when subject to root mean square error (RMSE) evaluation (Eq. (6)) [59]:

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^K (y_i - y_i')^2}{K}} \quad (6)$$

where  $y_i'$  is the experimental value,  $y_i$  is the theoretical value and  $K$  is the number of measurements.

### 3.2. Mass transfer correlations in microfluidic and millimetric parallel-plate electrochemical reactors

Mass transfer correlations were evaluated using electrochemical reactors equipped with  $d_{\text{elec}}$  of 100 and 500  $\mu\text{m}$  (representing micrometric setups) and 3000  $\mu\text{m}$  (representing a millimetric setup). The variation of mass transfer with fluid hydrodynamics, fluid physicochemical properties and electrochemical reactor geometry can be expressed using the theoretical dimensionless Sherwood ( $Sh$ ) correlation incorporating  $Re$ , Schmidt ( $Sc$ ), and dimensionless distance ( $Le$ ) numbers as well as the  $d_{\text{elec}}/W$  aspect ratio as given in Eq. (7) [60]:

$$Sh = a Re^b Sc^c Le^d \left( \frac{2}{1 + \frac{d_{\text{elec}}}{W}} \right)^{1/3} \quad (7)$$

The form of expression in Eq. (7) is valid with strict respect to the L ev eque approximation [57,61] which assumes that: (i) the electrolyte is incompressible, having constant physical properties; (ii) the flow is in laminar steady-state, becoming fully developed after a certain entry length for a given reactor geometry; (iii) mass transport is due to convection and diffusion; (iv) electrolyte flow is treated as bidimensional in the x-y planes (Fig. S1); (v) molecular diffusion of reactive species only occurs along the x-axis (normal to the electrodes); (vi) friction at walls induces retardation and produces a parabolic velocity profile with maximum velocity at the center of the channel; and (vii) the diffusional double-layer is a lot smaller than the hydrodynamic boundary layer. This dimensionless expression is particularly useful in estimating the average  $k_m$  values during scaling-up or scaling-down of an electrochemical reactor under the operating conditions of interest. The entry length ( $E_L$ ) was evaluated via Eq. (8) [61,62] and this increased from

0.027 to 21.7 mm when  $d_{\text{elec}}$  was increased from 100 to 3000  $\mu\text{m}$  (Fig. S3).  $Re$ ,  $Sc$  and  $Le$  are defined by Eqs. (9)–(11) whilst  $a$ ,  $b$ ,  $c$  and  $d$  are constant parameters.

$$E_L = 0.01 d_H Re \quad (8)$$

$$Re = \frac{u_L d_H}{\nu_L} \quad (9)$$

$$Sc = \frac{\nu_L}{D_L} \quad (10)$$

$$Le = \frac{d_H}{L} \quad (11)$$

$$d_H = \frac{2W d_{\text{elec}}}{W + d_{\text{elec}}} \quad (12)$$

$\nu_L$  is the kinematic viscosity in  $\text{m}^2 \text{s}^{-1}$  at the operating temperature [29,30,46] (Table S1).  $d_H$  is the hydraulic diameter in m.

To establish the theoretical  $Sh$  correlation,  $k_m$  values were measured at different electrolytic flow rates and temperatures. Experimental  $Sh$  values were computed using Eq. (13) [44] and plotted against various investigated  $Re$  and  $Sc$  numbers. The coefficients  $a$ ,  $b$ ,  $c$  and  $d$  were extracted from the best fit between theoretical and experimental  $Sh$  plots.

$$Sh = \frac{k_m d_H}{D_L} \quad (13)$$

The influence of electrolyte flow rate and temperature on  $k_m$  for each selected configuration is presented in Fig. 2.  $k_m$  increased with increasing flow rate across both configurations at constant temperature (Fig. 2(a)). This finding is expected owing to the enhancement of mass transport for the  $\text{Fe}(\text{CN})_6^{3-}$  reduction reaction when higher flow rates were tested. It can be observed that the increase in  $k_m$  with increasing electrolyte flow rate was much more pronounced in the microfluidic devices (100 and 500  $\mu\text{m}$ ) compared to the setup with a distance of 3000  $\mu\text{m}$  (Fig. 2(a)). The highest value of  $k_m$  ( $5.06 \times 10^{-5} \text{ m s}^{-1}$  at 0.05  $\text{L min}^{-1}$ ) was achieved when the electrochemical reactor was equipped with a 100  $\mu\text{m}$  interelectrode gap. In addition, when the temperature was increased from 10 to 50  $^\circ\text{C}$ ,  $k_m$  increased from  $3.28 \times 10^{-5}$  to  $5.22 \times 10^{-5} \text{ m s}^{-1}$  for the 100  $\mu\text{m}$  gap, from  $1.66 \times 10^{-5}$  to  $3.11 \times 10^{-5} \text{ m s}^{-1}$  for the 500  $\mu\text{m}$  gap and from  $1.20 \times 10^{-5}$  to  $2.41 \times 10^{-5} \text{ m s}^{-1}$  for the 3000  $\mu\text{m}$  gap (Fig. 2(b)). These trends are due to the increase in diffusivity of  $\text{Fe}(\text{CN})_6^{3-}$ -reducing species with temperature [46,63,64].

Using the  $k_m$  values obtained under different operating conditions, experimental values of  $Sh$  were obtained as illustrated in Fig. 3. The best fitting of Eq. (7) to the experimental data of each submillimetric and millimetric configuration was achieved when the coefficients  $a$ ,  $b$ ,  $c$  and  $d$  took the values provided in Table 1. The value of coefficient  $a$  varied slightly: it decreased from 3.00 to 2.31 from 100  $\mu\text{m}$  to 500  $\mu\text{m}$   $d_{\text{elec}}$  but increased again to 3.01 when using the 3000  $\mu\text{m}$  configuration. The values are within the range reported in the literature [60,65]. The variation of  $b$  between 0.32 and 0.37 corroborated the  $Re$  exponent value of close to 0.30 reported in the literature, which corresponds to fully developed laminar flow [34,37,60,65,66]. The widely reported 1/3 ratio of the  $Sc$  exponent  $c$  and the  $Le$  exponent  $d$  were obtained to match the experimental data under all three setups [30,32,34,37,60,65,66].

The fitting of the theoretical  $Sh$  correlations with the experimental data for all three  $d_{\text{elec}}$  configurations were evaluated using RMSE (Eq. (6)). Deviations of 0.038, 0.056 and 0.024 were calculated between logarithmic plots of the theoretical-experimental  $Sh$  values against  $Re$  for 100, 500 and 3000  $\mu\text{m}$  configurations, respectively. For  $Sc$ , the theoretical plot deviated from the experimental data by 0.070, 0.047 and 0.026 for gaps of 100, 500 and 3000  $\mu\text{m}$ , respectively. Therefore, the theoretical  $Sh$  correlations fitted very well with their experimental counterparts.

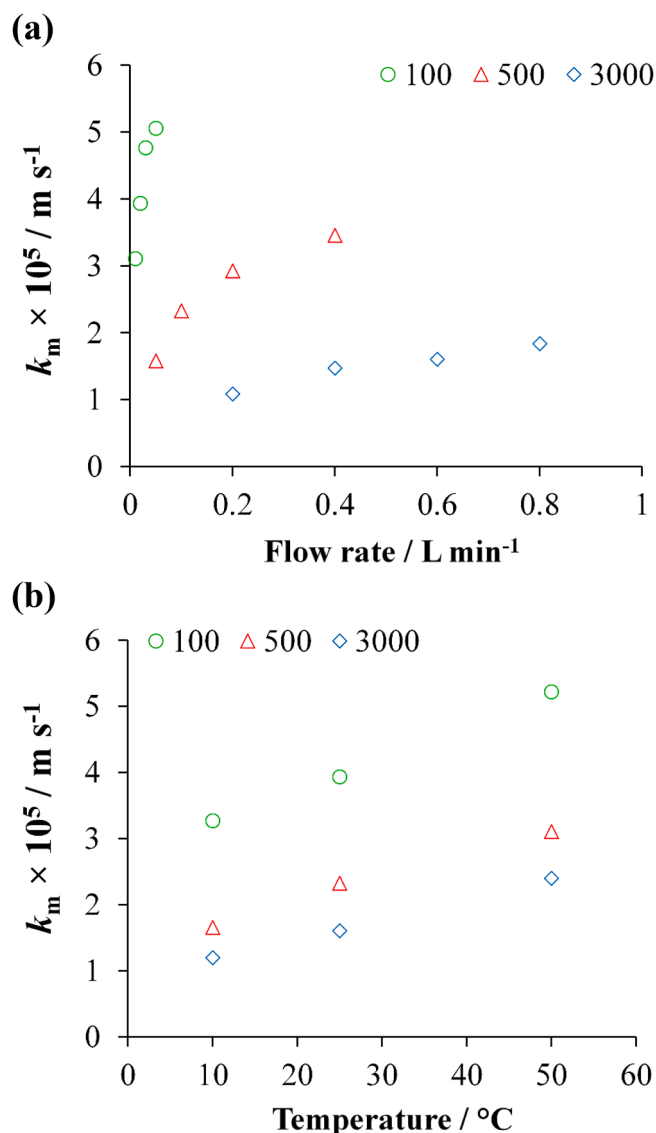


Fig. 2.  $k_m$  as a function of (a) electrolyte flow rate and (b) temperature for  $d_{\text{elec}}$  values of 100, 500 and 3000  $\mu\text{m}$ . Operating conditions: (a) temperature = 25  $^\circ\text{C}$  and (b) flow rates = 0.02, 0.10 and 0.60  $\text{L min}^{-1}$  for  $d_{\text{elec}}$  values of 100, 500 and 3000  $\mu\text{m}$ , respectively.

Given the trends in the  $a$ ,  $b$ ,  $c$  and  $d$  coefficients under the wide range of  $d_{\text{elec}}$  values studied, under a fully developed laminar regime, an average  $Sh$  correlation can be proposed (see Eq. (14)):

$$Sh_{\text{ave}} = 2.7733Re^{0.35}Sc^{1/3}Le^{1/3} \left( \frac{2}{1 + \frac{d_{\text{elec}}}{W}} \right)^{1/3} \quad (14)$$

The average  $Sh$  correlation is in close agreement with that proposed by Pickett et al. [60], with the  $a$  coefficient slightly higher in the current work. This could be attributed to the larger surface area used here (50  $\text{cm}^2$  vs. 16  $\text{cm}^2$  in [60]) as well as enhancement of mass transport in the microfluidic reactor.

#### 4. Conclusions

The variation in  $k_m$  as a function of  $d_{\text{elec}}$  from the millimetric to the micrometric range is reported for the first time. A novel single equation which estimates  $k_m$  over a wide range of  $d_{\text{elec}}$  values has been proposed. It highlights the inverse relationship between  $k_m$  and  $d_{\text{elec}}$ , where  $k_m$

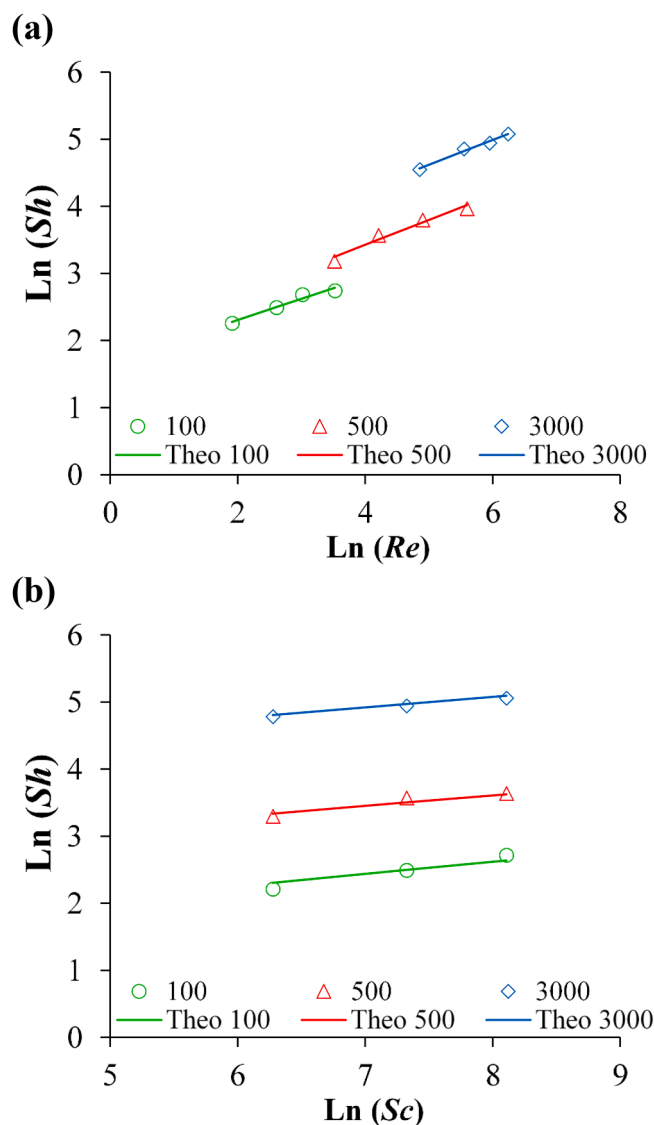


Fig. 3. Experimental (symbol) versus theoretical  $Sh$  correlations (continuous line) as a function of (a)  $Re$  and (b)  $Sc$  numbers. Operating conditions: (a) temperature = 25  $^\circ\text{C}$  and (b) flow rates = 0.02, 0.10 and 0.60  $\text{L min}^{-1}$  for  $d_{\text{elec}}$  values of 100, 500 and 3000  $\mu\text{m}$ , respectively.

Table 1

Fitted values of  $a$ ,  $b$ ,  $c$  and  $d$  parameters in the theoretical  $Sh$  correlation in parallel-plate electrochemical reactors with microfluidic and millimetric configurations under a laminar regime.

Configuration	$a$	$b$	$c$	$d$	$Re$ ranges	$Sc$ ranges
Thin film reactor (distance: 100 $\mu\text{m}$ )	3.00	0.32	0.33	0.33	7–34	532–3315
Thin film reactor (distance: 500 $\mu\text{m}$ )	2.31	0.37	0.33	0.33	34–269	532–3315
Parallel-plate (distance: 3000 $\mu\text{m}$ )	3.01	0.37	0.33	0.33	128–623	532–3315

values increase greatly at distances below 1000  $\mu\text{m}$ . Using experimental  $k_m$  data in submillimetric as well as millimetric configurations at varying flow rates ( $7 < Re < 623$ ) and temperatures ( $532 < Sc < 3315$ ), mass transport correlations have been established that take into consideration not only fluid properties but also electrochemical reactor hydrodynamics and geometry. The theoretical  $Sh$  number plots match very well

with their corresponding experimental values. The newly established *Sh* correlation in this work covers a wide range of interelectrode distances. The results were obtained using an electrochemical cell whose design is well adapted for multidisciplinary applications on a larger scale, whilst the robustness of the *Sh* correlation was justified experimentally. The results presented herein will be particularly helpful for  $k_m$  estimation in parallel-plate reactors under wide range of  $d_{elec}$  values (i.e. from several tens of micrometers to several millimeters) under given operating conditions. These expressions should be particularly suitable for micro-distances that can be useful for significant mass transfer intensification, unlike the millimetric or even centimetric distances often implemented.

#### CRedit authorship contribution statement

**Faidzul Hakim Adnan:** Methodology, Investigation, Formal analysis, Writing - original draft. **Marie-Noëlle Pons:** Supervision, Writing - review & editing, Funding acquisition. **Emmanuel Mousset:** Conceptualization, Methodology, Validation, Supervision, Writing - review & editing, Project administration, Resources, Funding acquisition.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.elecom.2021.107097>.

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