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Design principles of current collectors in microfluidic fuel cell with flow-through porous electrodes

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Abstract

Computational and experimental studies have been performed to investigate the effects of current collector design on the performance of microfluidic fuel cell (MFC) with flow-through porous electrodes. Characteristics of electron transport in MFC with flow-through porous electrodes are investigated based on a three-dimensional computational model. The lateral electron transport in the porous electrode is found to encounter high resistance. To improve the cell performance, influences of different current collector design parameters on the transport resistances are examined. Physical origins for the influences of different design parameters are also discussed. The results demonstrate that current collector position is the most influential factor due to the non-uniform flow rate distribution. In the experimental study, cell performances revealed maximum power density when current collectors were located at the high flow rate region. An increase of 61% was observed when the current collectors moved from the conventional exposed ends of the electrodes to the high flow rate regions in the electrode active area. Based on the results, some general rules are set for the current collector designs of MFCs.

Keywords: Microfluidic fuel cell; Porous electrode; Lateral electron transport; Electrical resistance; Current collector design

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>h</td>
<td>thickness in the z-direction, ( \text{mm} )</td>
</tr>
<tr>
<td>i</td>
<td>charge transfer current density, ( \text{A m}^{-2} )</td>
</tr>
<tr>
<td>L</td>
<td>length in the y-direction, ( \text{mm} )</td>
</tr>
<tr>
<td>( \sigma )</td>
<td>electrical conductivity, ( \text{S m}^{-1} )</td>
</tr>
<tr>
<td>( \phi )</td>
<td>potential, ( \text{V} )</td>
</tr>
<tr>
<td>A</td>
<td>center of the interface between external wire and current collector</td>
</tr>
<tr>
<td>B</td>
<td>center of the interface between current collector and electrode</td>
</tr>
</tbody>
</table>
1. Introduction

Microfluidic fuel cell (MFC) has received much attention in recent years as a promising alternative power source for small-scale electronic applications [1]. In MFC, the fuel and oxidant streams flow down a microchannel in a parallel manner and the inter-diffusive zone between these two streams is restricted to an interfacial width at the center of the microchannel [2]. Electrodes are integrated with sufficient distance from the inter-diffusive zone to prevent fuel crossover [3, 4]. Unlike conventional micro fuel cells which use physical barriers (mostly proton-exchange membranes), MFC utilizes the laminar nature of micro-channel flow to maintain the separation of the fuel and oxidant streams [5]. The unique membraneless feature helps avoid a series of membrane-related problems, such as water management, fuel crossover, membrane humidification, membrane degradation, etc. [6].

In this paper, a three-dimensional computational model for MFC with flow-through porous electrodes is developed with focus on the current collector design. Specifically, current collector and part of the external circuit, which are normally ignored in the previous modeling studies, are included in the present modeling domain. Effects of different current collector design parameters, including current collector size, positioning, thickness, electrical conductivity and connecting position of the external wire, are examined by parametric analysis and discussed. Experimental investigation on cell performances are also carried out to verify some of the theoretical findings: good agreement is achieved between the experimental results and the model predictions. Based on this analysis, some general design principles for the current collectors in MFC are derived, which can provide useful guidance for the future MFC development.

2. Numerical Model

2.1 Physical domain

The physical domain of the MFC studied in this work is shown in Fig. 1: three-dimensional diagram in Fig. 1a, x-y view in Fig. 1b and x-z view in Fig. 1c. Vanadium redox species in aqueous sulfuric acid solution are used in both half cells with $\text{V}^{2+}/\text{V}^{3+}$ as anolyte and $\text{VO}_2^2+/\text{VO}_2^{3+}$ as catholyte. Carbon paper is employed as the porous electrodes material. The rapid vanadium redox reactions on carbon electrodes eliminate the needs for any additional catalyst. In the operation, anolyte and catholyte enter the fuel cell via two opposite inlets of the anode and cathode, respectively. The two streams then pass through the inlet reservoirs, flow through the porous electrodes and meet at the common center channel. The redox reactions at the two electrodes are:

Anode: $\text{V}^{2+} \rightarrow \text{V}^{3+} + e^- \quad E^0 = -0.496 \text{ V vs SCE}$ (1)

Cathode: $\text{VO}_2^2+ + 2\text{H}^+ + e^- \rightarrow \text{VO}_2^{3+} + \text{H}_2\text{O} \quad E^0 = 0.75 \text{ V vs SCE}$ (2)

At the anode, oxidation reactions occur and electrons are generated. The generated electrons are drawn to the external circuit via the anode current collector and then transported to the cathode via the cathode current collector. At the cathode, reduction reactions take place and the electrons are consumed. When the two streams exit the electrodes into the common center channel, they will make a 90° turn and flow in a stratified, co-laminar format towards the outlet. The assumptions adopted in this analysis are:

1. Isothermal and steady state conditions;
2. Incompressible fluid flow;
3. Negligible effects of gravity;
4. Dilute solution approximation owing to the aqueous nature of the electrolyte;
(5) Negligible ionic migration due to the excess supporting electrolyte.

2.2 Computational procedure
The mass transport and charge conservation equations constrained by the boundary conditions are solved by the Finite Element Method (FEM) using the commercial software COMSOL MULTIPHYSICS Version 4.3. The segregated solver is used to accelerate the convergence since the coupling of the source terms between the species and charge transports makes the problem strongly non-linear. The following problem-solving procedure is adopted: the continuity and momentum conservation equations are first solved based on the initial setting. The results are consequently used for solving species and charge transports. A relative convergence tolerance of \(1 \times 10^{-4}\) is set. Structural meshes are generated and then a grid independence study is carried out on the base case. Three different grids are generated in the porous electrodes as shown in Fig. 2a. The cell sizes for Grid 1, Grid 2 and Grid 3 are 67\(\mu\)m\(\times\)83\(\mu\)m\(\times\)20\(\mu\)m, 50\(\mu\)m\(\times\)67\(\mu\)m\(\times\)15\(\mu\)m and 40\(\mu\)m\(\times\)55\(\mu\)m\(\times\)12\(\mu\)m (in x\(\times\)y\(\times\)z form), respectively.

2.3 Experimental validation
Validation of the model was conducted by comparing the numerically predicted cell performance with the experimental results in Ref. [7]. The geometric and operating conditions of the simulation follow the experimental study. Hydrophilic carbon paper (TGPH-060, Toray) was used as the porous electrode material. Stock electrolyte (50/50, V\(^{3+}/\)VO\(^{2+}\)) in 4 M aqueous H\(_2\)SO\(_4\) solution was recharged to produce V\(^{2+}\) and VO\(^{2+}\), the latter being the anolyte and catholyte, respectively. The electrode and electrolyte
properties are derived from Ref. [8,9]. Fig. 2b shows the comparison between the simulated and measured polarization curves of the cell. Good agreement is obtained between the simulated and experimental data.

3. Results and discussion

3.1 Modeling

In the analysis, rectangular-shaped current collectors are attached to the upper surfaces of the anode and cathode. Designs of the anode and cathode current collectors are set to be symmetric about the cell mid-plane (x = 0 mm) due to the symmetric flow pattern. Figures 3 and 4 show that the positioning of the current collector is a more influential factor than the size, due to the non-uniform velocity distribution.

3.2 Experiment

We carried out experiments to verify the results. An MFC with flow-through porous electrodes, as shown in Fig. 5, was fabricated. Three pairs of ports (marked with A1, A2, B1, B2, C1, C2) were simultaneously planted at different positions along the porous electrodes (Fig 5 (b) and (c)) in a single cell to eliminate the deviations from cell fabrication. In the experiment, the current collector size was kept unchanged (L = 1 mm, W = 1 mm, h = 5 mm) while the position of the current collector center varied from y_B = -7.5 mm to y_B = 11.5 mm. This was realized by connecting the external circuit to the cell via the port-pairs A1-B1, A2-B2, and A3-B3, respectively. Vanadium species based redox electrolyte was fed into the cell at different flow rates. The polarization curves were recorded with a potentiostat, as shown in Fig. 6. The experimental results exhibited trend consistent with the modeling results.
3.3 Current collector design

The validated numerical model was employed for current collector design analysis. The results are presented in Fig. 7. As shown in Fig. 7a, when the thickness decreases from $3 \times 10^{-1}$ mm to $1.5 \times 10^{-1}$ mm, no evident change in the polarization curves is observed. When the thickness decreases to $4 \times 10^{-4}$ mm, a 16% drop in maximum output power occurs. At this condition, the lateral electron transport in the current collector has no apparent priority over that in the porous electrode, electrode potential becomes non-uniform along the y-direction, electrochemical reaction rate decreases, especially in the high flow rate region and accordingly the cell performance diminishes.

The effect of the electrical conductivity of the current collector material is shown in Fig. 7b. The upper bound ($1 \times 10^7$ S m$^{-1}$) represents the magnitude of typical conductivity values for conductive metallic current collectors (Au, Ag, Cu, etc.), while the lower bound ($5 \times 10^3$ S m$^{-1}$) for the carbon based materials. Metallic current collectors can significantly reduce the electrode potential loss due to the lateral electron transport in the porous electrode.

Normally, potential distribution within the current collector is uniform due to the high electrical conductivity. In case of low conductivity and/or small thickness, non-uniform potential distribution may occur within the current collector. As a result, the connecting position between the external wire and the current collector becomes critical as the electron transport in the z-direction towards the external circuit will be more concentrated to the region covered by the external wire. Effects of the external wire positioning are examined and the results are presented in Fig. 7c for the cell with a low current collector conductivity ($5 \times 10^3$ S m$^{-1}$). An increase of 18.9% in the maximum power is observed when we move the external wire position along the electrode midline from $y_A = -7$ mm to $y_A = 11$ mm. Thus, in the cases with a low current collector conductivity or a small current collector thickness, effects of the current collector are weakened and external wire plays the role of the current collector alternatively. To reduce the electrical resistance, the external wires need to be attached to the high flow rate region.
4. Conclusions  
From the investigations above, some basic principles in the current collector design for MFCs can be drawn:  The most important point is that the current collector should be located at the high flow rate region; As for the size of the current collector, as long as it can cover the high flow rate region and be thick enough (>10^{-3} mm), there should be no further need to increase the size as it won’t significantly improve the output; The conductivity of the current collector material should be larger than the electrode material, and highly conductive metallic materials are preferred; The wiring position is insignificant if the above criteria are met, otherwise, if low conductivity material has to be used and/or the thickness is limited, the external wire should also be attached to the current collector at the high flow rate region. These findings can provide useful guidance for the future development and manufacturing planning of MFC.
Biography

Prof. Michael K.H. Leung is an Associate Dean in the School of Energy and Environment and the Director of Ability R&D Energy Research Centre at the City University of Hong Kong. His research interests include fuel cell and photocatalysis.