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Numerical and experimental comparative study of microfluidic fuel cells with different flow configurations: Co-flow vs. counter-flow cell

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HIGHLIGHTS

- Comparative study of co-flow and counter-flow microfluidic fuel cells is conducted.
- The numerical models and experimental prototypes fit very well with each other.
- The counter-flow cell exhibits uneven current distribution and poorer performance.
- The counter-flow cell achieves much higher fuel utilization than the co-flow cell.
- Design principles are suggested for counter-flow cells to improve the performance.

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ABSTRACT

Microfluidic fuel cell (MFC) is a new type of fuel cell which utilizes two laminar flows as electrolyte for both reactant delivery and ionic conduction purposes. According to the flow configuration, two MFC designs, that is, the co-flow cell and the counter-flow cell, have been proposed in the literature, but the specific merits and demerits between them have not been well understood yet. Therefore, in this work both experimental and numerical comparative studies are conducted on this issue. It is found that the counter-flow MFC encounters an inherent drawback on its power output because the outer part of its electrodes contributes much less than the inner part due to the longer ionic transport path. In conclusion, short and wide electrodes are more appropriate for counter-flow MFCs rather than the conventional long and narrow ones. Nevertheless, the counter-flow MFC does exhibit great advantage on low flow rate tolerance, leading to much improved fuel utilization and energy density at the same time. Considering this huge superiority for real applications, structural optimization of the counter-flow MFC is further conducted by reducing the electrode distance and discarding the outer part of the electrodes. The optimized counter-flow cell performance is successfully improved to the same level with its co-flow counterpart.

1. Introduction

Fuel cell is very promising as a future power source because of its distinct characteristics such as high energy efficiency, superior energy density, environmental friendliness, long-term energy storage and generation ability. Various applications of this technology are emerging in recent years, especially in the field of transportation and stationary power generation where high rated power is required. As for the market of small-power applications, the competitiveness of fuel cell is greatly impaired by its relatively high cost compared with other existing technologies such as batteries. To lower down the cost, a novel-type microfluidic fuel cell (MFC) has been proposed recently [1,2], which utilizes the characteristics of laminar flow for fuel-oxidant separation purpose instead of the conventional solid membrane. As shown in Fig. 1(a), a typical MFC employs two laminar flow streams, one mixed with fuel (anolyte) and the other mixed with oxidant (catholyte), to flow in parallel along the micro-channel. In this manner, the high-cost membrane in conventional fuel cells can be eliminated together with its dry-out and degradation issues [3,4]. In addition, the reaction heat and water generated can be efficiently removed by the flowing electrolyte, leading to a much simplified balance of plant (BOP). As a consequence, the fabrication cost of MFC is much lowered, which makes it especially suitable as mini-watt power sources.

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Nomenclature

Symbol

ρ	density (kg m^{-3})
u	velocity (m s^{-1})
P	pressure (Pa)
μ	dynamic viscosity (Pa·s)
ω	mass fraction
x	molar fraction
j	diffusion flux (kg m^{-2} s^{-1})
S	production/consumption rate due to electrochemical reactions (kg m^{-3} s^{-1})
M	molar mass (kg mol^{-1})
D	diffusion coefficient (m^2 s^{-1})
c	molar concentration (mol m^{-3})
M_n	average molar mass of the mixture (kg mol^{-1})
σ	conductivity (S m^{-1})
φ	potential (V)
i	current density (A m^{-2})
ν	stoichiometric coefficient
n	number of transfer electrons
F	Faraday's constant (96,485 C mol^{-1})
i_0	exchange current density (A m^{-2})
χ	reaction order
α	charge transfer coefficient
η	activation overpotential (V)
R	gas constant (8.314 Jmol^{-1} K^{-1})
T	temperature (K)
E_{eq}	equilibrium potential (V)
Q	charge source term (A m^{-3})

Subscripts

a	anode
c	cathode
f	fuel
i, j, k	species
l	electrolyte
m	main reaction
o	oxidant
p	parasitic reaction
s	electrode
0	standard, reference, or boundary value

Superscript

$x \rightarrow 1$	infinite solution

To date, there are plenty of MFC prototypes proposed in the literature, which have greatly contributed to the research and development of this technology. Most of these prototypes utilize the co-flow configuration as shown in Fig. 1(a), in which the diffusion direction of reactant crossover is orthogonal to the flow direction of electrolyte solution. This configuration has been the mainstream ever since the MFC technology was proposed, probably because of its simple structure and easy fabrication. During the cell operation, the convective electrolyte flow is much faster than the diffusive reactant crossover (that is, a high Peclet number), which can restrict the mixing zone of fuel and oxidant (as indicated by the white dash line in the figure) to a thin layer in the middle of the channel. In this manner, both the anode and cathode can work well so the co-flow MFC can generate very high power output with various types of fuels, some of which are even comparable to the PEMFC. Kjeang et al. [5] developed an all-vanadium co-flow MFC which can obtain a peak power density of 70 mW cm^{-2} at room temperature. Lu et al. [6] employed the dual-electrolyte technique in their hydrogen-oxygen co-flow MFC with acid electrolyte for the cathode side and alkaline electrolyte for the anode side. A peak power density as high as 1.3 W cm^{-2} was achievable together with a high OCV of 1.89 V. Hollinger et al. [7] proposed a direct methanol co-flow MFC which can generate a power output of 70 mW cm^{-2} and a current output of about 650 mA cm^{-2}. As for the formic acid fuel, a peak power density of 55 mW cm^{-2} was achieved by Jayashree et al. [8] using 1 M formic acid and 50 sccm oxygen. These high power outputs are mainly attributed to both the sufficient supply of reactant to the electrode surface and the effective suppression of reactant crossover, which generally requires a relatively high electrolyte flow rate. Nevertheless, the fuel utilization is consequently sacrificed due to the limited retention time of the fuel inside the channel. Moreover, a large amount of solution needs to be stored inside the co-flow MFC system, leading to a low energy density. These shortcomings will greatly impair its application potential. In addition to the co-flow configuration, the counter-flow configuration as shown in Fig. 1(b) has also been proposed, in which the diffusion direction of fuel and oxidant crossover is opposite to the flow direction of catholyte and anolyte, respectively. This kind of MFC is less studied in the literature, although the fabrication process is as easy as its co-flow counterpart. When coupled with 3D flow-through electrodes, the counter-flow MFC can also achieve satisfactory power output. Ibrahim et al. [9] developed an all-vanadium counter-flow MFC and obtained a peak power density of 760 mW cm^{-2}. Gurrola et al. [10] utilized 3 M formic acid as fuel and the achieved peak power density was as high as 100 mW cm^{-2}. As for the ethanol fuel, a peak power density of 99.4 mW cm^{-2} was generated by a dual-electrolyte counter-flow MFC developed by López-Rico et al. [11] Furthermore, the counter-flow MFC is considered to be highly tolerable to low electrolyte flow rates without the detrimental effect from aggravated reactant crossover. This is because that the convective electrolyte counter-flow can effectively prevent the diffusive reactant crossover from the electrode surface. As a consequence, high fuel utilization and low electrolyte consumption may be achievable by counter-flow MFCs. However, the cell power output may be sacrificed due to the insufficient reactant supply.

In addition to experimental approaches, numerical modeling is also an effective method for MFC study, which cannot only reveal the in-depth mechanisms behind the experimental phenomenon but also optimize the cell performance economically. A number of MFC modeling studies have already been published in the past decade, most of which focused on the co-flow configuration while only a few of them investigated the counter-flow configuration. For the co-flow MFC, great efforts have been made on the improvement of its fuel utilization by using various strategies, such as the increase of channel aspect ratio [12–14], the utilization of tapered electrodes [12] or very small electrodes [15], introduction of a third electrolyte [14,16,17], using multiple inlets [18], employing flow-through porous electrodes [19] and using novel channel geometries [20]. As for the counter-flow MFC, Xuan et al. [21] introduced micro-ridges into the microfluidic channel in order to generate chaotic flows, which can dramatically improve the cell current and power output. Xu et al. [22] investigated the possibility of counter-flow MFCs with low flow rate operation. With 1 M formic acid as fuel, the highest fuel utilization of 91.4% was achieved at 1 μl min^{-1}. Other modeling studies focused on counter-flow