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Performance analysis of microfluidic fuel cells with various inlet locations and multiple compartments

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ABSTRACT

Computational evaluation of three design modifications of a membraneless microfluidic fuel cell (MMFC), i.e., inlets located midway along the microchannel, multiple compartments in the channel cross section, and a multi-stream (oxidant-fuel-oxidant) configuration, is performed in this work. The first two modifications are novel concepts proposed in this work. These adjustments to the microchannel are introduced to decrease the negative effects caused by the increased channel length, width, and height on the performance of MMFCs. Formic acid and oxygen are dissolved in sulfuric acid solution as the fuel and oxidant, respectively. Simulations are executed by employing three-dimensional Navier-Stokes equations and mass transport equation for the analyses of flow and species concentration. The electro-chemical reaction is modeled using Butler-Volmer equations. Based on the study that investigated the inlet location, inlets placed at the center of the microchannel are shown to be the best option among the selected inlet positions. Additionally, the multi-compartment configuration enhances the current density by as much as three times compared to a simple square microchannel.

1. Introduction

Fuel cells are electrochemical devices that perform oxidation and reduction reactions between the fuel and oxidant at the anode and cathode to produce electricity [1–3]. It is predicted that conventional battery technologies will not be able to meet the increasing energy demands for modern portable devices, i.e., cell phones, global positioning systems (GPSs), remote sensors, and *in vivo* smart diagnostic medical devices [4], which require extended battery periods without recharging [5]. However, fuel cells are anticipated to be a solution to this problem.

Dyer [4] suggested that fuel cells have the advantage of higher power densities compared to Li-ion batteries. Miniaturization of fuel cells could further increase the power density because miniaturization increases the surface-to-volume ratio, thereby promoting reactions [4,5]. However, several mechanical and technological difficulties have been reported while attempting to miniaturize fuel cells [6–12]. These difficulties are caused by membrane-related ohmic overpotential, fabrication of bipolar plates made of graphite, and management of heat and water [10–15]. Therefore, to overcome these difficulties, miniaturization of fuel cells for high power density applications must be performed with novel fuel cell architectures.

Membraneless microfluidic fuel cells (MMFCs) separate fuel and oxidant streams by taking advantage of the nature of laminar flow [16]; thus, they are also called “co-laminar flow fuel cells.” By removing the membrane, MMFCs can avoid several membrane-related problems such as ohmic losses, fouling problems, and sealing and water management problems [15–18]. MMFCs also have the advantage of using liquid fuels (instead of gaseous fuels), which have higher power densities. This is important for producing high power outputs for portable power applications [16,19].

The performance of MMFCs is sensitive to the microchannel geometry because the diffusion process between the laminar streams in MMFCs is affected by the channel geometry. In MMFCs, a mixing region is created between the two streams of reactants for diffusive mixing and develops along the channel. This mixing region reduces the availability of the electrodes to the reactants and can be affected by the cross-sectional shape of the microchannel. Tapering the electrode enlarges the active surface area of the electrode. However, there is a limit to tapering of the electrodes because crossover issues arise beyond a certain limit. A depletion layer is formed on each electrode, preventing the reactants from reaching the electrode; reducing the thickness of the depletion layer is desirable [5].

Choban et al. [20] proposed an MMFC that employed formic acid (fuel) and oxygen (oxidant). They demonstrated that two parallel laminar streams underwent negligible mixing with each other; thus, there was no need to use a membrane to separate the streams in the fuel cell. Choban et al. [19] investigated a Y-shaped MMFC that produced a

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Nomenclature

$E_{REVERSIBLE}$	reversible potential (V)
F	Faraday constant (C mol^{-1})
R	general gas constant (J mol^{-1} K^{-1})
P_i	net rate of production (mol m^{-3} s^{-1})
D_i	diffusion coefficient (m^2 s^{-1})
Y_i	mass fraction of species
i	reaction rate (A mol C^{-1} m^{-3})
U	average flow velocity (m s^{-1})
y	distance along channel from inlet (m)
N	number of cells
C_i	species concentration
S	rate of reactant species' consumption
p	absolute pressure (Pa)
a	density of catalyzed active area
i_0	exchange current density (A m^{-2})
n	number of electrons involved in the reaction
T	absolute temperature (K)
e_i	mole number of species (mol m^{-3})
L_{in}	inlet location (m)
L	channel length (m)
W	channel width (m)
H	channel height (m)
W_a	anodic channel thickness (m)
W_c	cathodic channel thickness (m)
W_{ext}	width of electrode extended toward center of channel (m)
E_a	width of anode electrode for oxidant-fuel-oxidant configuration (m)

Greek symbols

Φ	electrical potential (V)
σ_i	electrical conductivity (S m^{-1})
σ	variance
ρ	density (Kg m^{-3})
μ	dynamic viscosity (Pa s)
α	charge transfer coefficient
η	activation overpotential (V)
ϕ_s	solid phase potential difference (V)
ϕ_e	electrolyte phase potential difference (V)

current density of 0.4 mA cm^{-2}. They used an acidic solution to minimize the resistance to transportation of protons in the channel. Different geometries of T-shaped MMFCs were studied for effective fuel utilization by Bazylak et al. [5]. They found that a square channel showed better performance than a channel with a rectangular cross section; additionally, tapered electrodes could be employed to enhance fuel utilization. Chang et al. [21] investigated an MMFC by varying the flow rate, Péclet number, and concentrations of fuel and oxidant to improve the performance of the MMFC. They suggested that, to avoid fuel crossover, the MMFC should be operated at a high Péclet number. Additionally, increasing the cathodic stream concentration was more effective for enhancing the power than was increasing the anodic stream concentration.

The fuel crossover issue arises from complete mixing of two streams. Khabbazi et al. [22] investigated MMFCs with multi-stream inlets, multiple periodically-placed inlets, and a tapered channel with different aspect ratios of the channel and electrode geometries. They suggested that multiple periodically-placed inlets effectively filled the depletion region, and that tapering the channel enhanced fuel utilization. Salloum et al. [23] used porous electrodes and vanadium redox species as the oxidant and fuel in a convective counter-flow MMFC. A stream of sulfuric acid was used as a non-reacting electrolyte for conduction of cations and to maintain separation between both streams to avoid diffusive mixing. The electrolyte was injected at the center of the MMFC to prevent the streams from merging. Preventing the oxidant and fuel from undergoing diffusive mixing in the counter-flow MMFC allowed them to be reused. They found that increasing the flow rate increased the power output but also resulted in substantial losses in fuel utilization. They achieved 24.9% fuel utilization at 50 μL min^{-1}.

Lopez-Montesinos et al. [24] developed a three-dimensional numerical model of an MMFC where the main channel had a bridge-shaped cross section. They found that utilizing a microchannel with a bridge-shaped cross section minimized the mixing region thickness, enhanced reactant utilization, and improved the performance of the MMFC. They suggested that, in order to achieve optimal performance, the flow rate needed to be adjusted in accordance with the concentration of the oxidant being used. By testing bridge structures with various aspect ratios, they found that a lower aspect ratio was more desirable for this structure. Sun et al. [25] investigated an MMFC that used multi-stream laminar flow by introducing a third stream (of an electrolyte) to separate the oxidant and fuel and to prevent them from reacting or undergoing diffusive mixing. They found that a third stream of sulfuric acid accelerated the transport of ions; additionally, the third stream could be controlled to improve the performance of the MMFC. Lee and Ahn [26] proposed MMFC single stacks, built in series and in parallel, that utilized multiple laminar flows to build multi-stream MMFCs. They built multi-stream MMFCs in a single stack comprised of one channel.

In the authors' previous work [27], a Y-shaped MMFC was investigated with various geometric modifications. Ten different channel cross-section configurations were tested, and the best model was suggested. It was found that decreasing the channel length, height, and width led to increased power density. Wang et al. [28] studied an MMFC for different flow configurations, i.e., parallel flow and counter flow, to investigate their merits and demerits numerically and experimentally. They found that counter-flow MMFC performed poorly and unevenly as compared to parallel-flow MMFC, and carried out structure optimization to improve the performance of the counter-flow MMFC. Wang and Leung [29] proposed a circular six cell stack connected in series to improve the efficiency and power density of an MMFC. They found that the stack performance is not affected by flow rate of electrolyte. With the stack, they were successful to achieve 108.7 mW cm^{-2} peak power density. Yang et al. [30] created an array of fuel cell based on laminar flow for portable lab-on-chip devices. To scale-up the performance of the MMFC, they used multiple branched Y-shaped channels in a single microfluidic chip. With the array incorporating four fuel cells connected in series, they achieved 60.5 μW cm^{-2} power density.

As described above, miniaturization has also contributed to higher power densities in devices. Miniaturization can be achieved by eliminating the membrane and exploiting the nature of laminar flow. However, additional efforts are required to improve the performance and fuel utilization of MMFCs. Previous studies [5,16,22,27] found that increasing the channel length, width, and height decreased the performance, while reducing these parameters decreased fuel utilization. The diffusive mixing region at the liquid-liquid interface and the depletion region around the electrodes act to decrease both performance and fuel utilization [5,16,22]. Many geometric modifications [5,16–34] for the channel and electrodes have been proposed to improve performance and fuel utilization. The present study represents another experiment in this line of research.

Herein, three methods were investigated to alleviate the negative effects of increased channel length, width, and height on the performance of MMFCs. The fluid flow, species concentration, and reaction kinetics in the MMFC were numerically modeled using three-dimensional Navier-Stokes equations, convection and diffusion equations, and