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Performance and design analysis of tubular-shaped passive direct methanol fuel cells

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ABSTRACT

To achieve the maximum performance from a Direct Methanol Fuel Cell (DMFC), one must not only investigate the materials and configuration of the MEA layers, but also consider alternative cell geometries that produce a higher instantaneous power while occupying the same cell volume. In this work, a two-dimensional, two-phase, non-isothermal model was developed to investigate the steady-state performance and design characteristics of a tubular-shaped, passive DMFC. Under certain geometric conditions, it was found that a tubular DMFC can produce a higher instantaneous Volumetric Power Density than a planar DMFC. Increasing the ambient temperature from 20 to 40 °C increases the peak power density produced by the fuel cell by 11.3 mW cm^{-2} with 1 M, 16.3 mW cm^{-2} with 2 M, but by only 8.4 mW cm^{-2} with 3 M methanol. The poor performance with 3 M methanol at a higher ambient temperature is caused by increased methanol crossover and significant oxygen depletion along the Cathode Transport Layer (CTL). For a 5 cm long tubular DMFC to maintain sufficient Oxygen transport, the thickness of the CTL must be greater than 1 mm for 1 M operation, greater than 5 mm for 2 M operation, and greater than 10 mm for 3 M or higher operation.

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1. Introduction

As the world-wide demand for energy continues to increase, the importance of finding new sources of energy with higher efficiencies and less overall impact on the environment will continue to grow exponentially. A fuel cell is an electrochemical energy conversion device that takes energy, provided by fuel (hydrogen, alcohol, hydrocarbons, etc.) and an oxidant (oxygen, air, chlorine, etc.), to produce a current of electrons, i.e. electricity. There are several different types of fuel cells, but they all are composed of a similar design consisting of two electrodes (a negative anode and a positive cathode) separated by an electrolyte. In some fuel cells, the electrolyte conductively separates the anode and cathode while allowing the transport of electrically charged particles (protons) across its surface. Specifically, this paper focuses on the Direct Methanol Fuel Cell (DMFC), which is a type of Polymer Electrolyte Membrane (PEM) Fuel Cell.

As a direct result of recent developments in portable, hand-held technology that demands larger outputs in power density and new, alternate sources of energy, the DMFC has gained a lot of attention due to its higher energy density compared to Lithium Ion batteries. The DMFC operates at low temperatures, gives off low emissions, is based on a simple geometry that can provide completely passive energy, and methanol, its fuel, provides high energy content along with safe storage capabilities [1]. There are two basic half-reactions that occur in a DMFC:

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- Liquid methanol and water are broken into protons, electrons, and carbon dioxide gas at the Anode Catalyst Layer (ACL):

$$CH_3OH + H_2O \rightarrow 6H^+ + 6e^- + CO_2 \quad (1)$$

- Oxygen gas, protons, and electrons are converted into water at the Cathode Catalyst Layer (CCL):

$$\frac{3}{2}O_2 + 6H^+ + 6e^- \rightarrow 3H_2O \quad (2)$$

- The overall reaction is:

$$CH_3OH + \frac{3}{2}O_2 \rightarrow 2H_2O + CO_2 \quad (3)$$

There are three main types of DMFCs: active, semi-passive, and passive. An active DMFC requires external pumps and fans to force the flow of methanol and oxygen to the fuel cell. This type of DMFC produces the highest power density (as a result of always having sufficient fuel or oxidant in the catalyst layers), but the addition of auxiliary components (pumps, fans, heaters, external humidity equipment) reduces the overall theoretical efficiency of the fuel cell since they require electricity to run. A semi-passive DMFC operates with an active anode and passive cathode or a passive anode and active cathode. On the other hand, a passive DMFC operates completely on its own. Methanol is mainly transported to the ACL due to concentration gradients and gravity which force the fuel through porous diffusion layers (Anode Diffusion Layer (ADL) and Anode Micro-Porous Layer (AMPL)) that provide mass transport resistance, while oxygen is readily provided from the air and transported by natural convection and diffusion to the CCL.

Previously, most DMFC research efforts were focused on planar, flat geometries. This work considers a tubular DMFC geometry that has the anode on the inside of the tube and the cathode on the outside of the tube. Considering a fuel cell that produces the same voltage and current density, independent of the geometry, which is a valid assumption if the same Membrane Electrode Assembly (MEA) and fuel are used to build and operate both a tubular and a planar DMFC; a tubular-shaped fuel cell has several distinct advantages over a planar-shaped fuel cell:

- For certain geometric conditions, the tubular DMFC has a higher instantaneous Volumetric Power Density (VPD) than the planar DMFC associated with higher instantaneous power output per unit volume.
- Ability to operate in all orientations without lack of methanol contacting the ACL.
- Reduced cost due to decreased volume of materials.
- Same shape as existing AA, AAA, D, and C batteries, which allows easier conversion between batteries and fuel cells in the future.
- With the elimination of flow fields at the cathode, uniform pressure is applied across the membrane electrode assembly (MEA) [2].

Based on these significant advantages, the passive, tubular DMFC should be considered a potential replacement for the passive, planar DMFC, which is currently aggressively investigated.

There have been very few experimental [3–11] and numerical [12] research efforts on tubular-shaped DMFCs. Considering the experimental work conducted on the tubular-shaped DMFCs, the main challenge has been to construct the fuel cell in such a way that prevents leaks and reduces the internal resistance. Kunimatsu and Okada [3] developed a semi-passive (active anode and passive cathode), tubular DMFC operating with 1 M methanol solution that produced 12 mW cm^{-2} with methanol pumped through the anode channel at 1 ml h^{-1}. They found that the catalyst layer composition and hot-pressing process were crucial to improving the performance of tubular-shaped DMFCs. Qiao et al. [4] used a wet-chemical deposition process to coat the anode catalyst layer onto a tubular membrane made from Flemion tubing. During half-cell testing, the micro-tubular DMFC produced 1.8 mW cm^{-2} with a 2.8 mg cm^{-2} anode catalyst loading. Qiao et al. [5] further developed a method to coat the catalyst onto the membrane of a tubular DMFC by using an impregnation reduction method. They [6] also created a process by which the chemical reduction of Pt could be used to deposit a cathode catalyst layer onto the tubular membrane of a DMFC. They were able to carefully control the loading and thickness of the catalyst layer.

Shao et al. [7,8] built a tubular DMFC with a Titanium mesh current collector at the anode and cathode. They developed a method of dipping the Titanium mesh into different solutions until desired loads of Nafion® layers, catalyst layers, and Gas Diffusion Layers (GDL) were achieved. They explained this procedure and its results during half-cell testing of the anode and cathode. M.S. Yazici [9] developed a tubular fuel cell that could operate with Hydrogen or Methanol at the anode and used a material called GRAFCELL® (porous graphite material) as the GDL to control the fuel flow rate to each catalyst layer. Different open-hole ratios of graphite GDL material were investigated and it was concluded that water and air management at the cathode, as well as, liquid fuel management at the anode could be accomplished with different pore sizes and thinner GDLs.

Yu et al. [10] developed a semi-passive tubular DMFC that produced 10 mW cm^{-2} with 4 M methanol solution flowing through the anode at 80 °C. Their DMFC included a new electrolyte membrane made from a porous silica pipe that had pores filled with perfluorinated resin and analyzed the membrane using a scanning electron microscope (SEM), electrochemical impedance spectroscopy system (EIS), and the bubble method to analyze the cross sections, conductivity, and porosity, respectively. Lee et al. [11] investigated the advantages of developing a tubular versus a planar DMFC including the overall specific power and volumetric energy density. They also constructed a tubular DMFC that consisted of 6 small planar DMFCs connected in a circular design that allowed passive anode and cathode operation that produced 12 mW cm^{-2} with 3 M methanol solution at room temperature. Most recently, Ward et al. [12] designed and built a tubular-shaped fuel cell that operated completely passively. By constructing the fuel cell with a tubular-shaped frame, both Nafion® 212 and 115

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