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Model-based analysis of thermal and geometrical effects in a microscale methanol fuel cell

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ABSTRACT

This paper reports the development of a mathematical model to predict the performance of a microscale methanol fuel cell with a single fuel/electrolyte channel. Performance of the cell is investigated as a function of fuel stream inlet temperature and catalyst deposition geometry. The model is fit to experimental data by maximizing the coefficient of determination, R^2. Results show that peak power density with regard to total exposed catalyst surface area is inversely proportional to catalyst deposition width and proportional to fuel stream temperature. For both parameters, the mathematical model was found to compare well with experimental results in the operating regime preceding and including maximum power density. The model presented here can be used to optimize these parameters during the design phase.

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Introduction

The increase in the number of smart devices, the high capacity requirement of electronic devices, and the desire to recharge quickly are driving the portable power market. These growing demands have led to the investigation of microscale fuel cells as an alternative to current battery technology [1–6]. Particularly in areas where connectivity to the main power grid is not possible, fuel cells can provide instantaneous power restoration [7]. Furthermore, microscale fuel cells demonstrate superior energy densities and the potential to provide longer off-grid run times than current battery technology [8]. Several microscale fuel cell architectures have been proposed to address this growing demand for portable power. Microscale fuel cells that are referred to as microfluidic fuel cells, membraneless fuel cells, or laminar-flow-based fuel cells commonly employ separate fuel and electrolyte streams and operate without a membrane separating the anode and cathode. In the work presented here, the microscale fuel cell utilizes a membrane hot-pressed to the cathode and a single fuel/electrolyte stream. In microscale fuel cells having a single inlet, a single outlet, and a relatively high aspect ratio, defined as the ratio of fuel flow chamber length to width, fuel depletion resultant from slowly circulating flow patterns adjacent to the quickly convecting central stream may appreciably reduce average power density [9–11].

To address this mass transport limitation, several studies have explored strategies for maintaining a high reactant concentration near electrodes. A study by Yoon et al.

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demonstrated that multiple inlets or outlets can be utilized to remove depletion boundary layers and thereby replenish reactants to flow-over electrodes [12]. As an alternative to flow-over electrodes, Kjeang et al. suggested the use of flow-through, porous electrodes to decrease the influence of boundary layer depletion [13–15]. More recent studies show that shorter and wider electrodes (low aspect ratio) can reduce reactant depletion at electrode surfaces, thus improving reaction kinetics and current densities [16,17]. Modeling efforts show that other geometric modifications, such as tapering of channels and electrodes, can also be used to reduce mass transport losses [18,19]. However, a recent review by Goulet et al. highlights the need for a combined modeling and experimental approach to evaluating electrode design in a microscale fuel cell [3]. To this end, we have developed a model which is fit to experimental data. Comparisons between different aspect ratio electrodes (ratio of fuel chamber length to width) show that low-aspect-ratio electrodes achieve more than double the peak power densities of high-aspect-ratio electrodes [20].

Here we show an improved mathematical model, validated with experimental data, to investigate the performance of a low aspect ratio microscale methanol fuel cell. The model is improved via refinement of the semi-empirical governing equation [20]. Statistical analysis is added to provide a measure of goodness-of-fit between the modeling and experimental data. In addition, performance is evaluated as a function of catalyst deposition geometry and fuel inlet stream temperature as a means to investigate directly simulated and non-simulated parameters, respectively. Most microscale fuel cells reported to date are designed to have catalyst deposition length and width identical to fuel chamber length and width. Length is defined as the direction parallel to the inlet direction, and width is perpendicular to the inlet direction. Here we investigate catalyst deposition geometries that are different from the fuel chamber geometry (Fig. 1). Prior models of microfluidic fuel cells have not investigated catalyst deposition geometry [16,18,19]. In addition, these models study microscale fuel cell architectures in which two streams (fuel and oxidant) flow between electrodes. In the model presented here, electrodes are separated by a membrane and a single stream (fuel/electrolyte). Furthermore, this model evaluates a fuel chamber with a width greater than its length (low aspect ratio), based upon previous electrode design rules [17]. Prior modeling efforts evaluate fuel chambers with a length substantially greater than its width (high aspect ratio) [16,18,19]. The results presented here can further the development of microscale fuel cells as portable power sources by allowing for operating parameters to be optimized before fabrication and testing in an experimental setup.

Materials and methods

Electrode preparation

Several catalyst deposition geometries (active catalyst area) were considered. For all studies, anodes utilized 1.5 mg/cm^2 Pt/Ru/C catalyst (Alfa Aesar, 50:25:25 wt%) and cathodes utilized 1.5 mg/cm^2 Pt/C catalyst (Alfa Aesar, 50:50 wt%). Nafion binder (5 wt% solution, Ion Power Inc.) was added to the catalysts at a 5:3 wt ratio of catalyst to binder. Catalyst inks were prepared by dispersing the catalyst and binder in a 1:1 (V/V) solution of H_2O:2-propanol. The inks were sonicated (Sonics VCX-130PB) for 5 min to ensure uniform mixing and were then hand-painted onto the microporous side of Sigracet 25BC carbon paper (FuelCellsEtc).

Nafion 212 (FuelCellsEtc) was bonded (Carver 5420) to each cathodic gas diffusion electrode (GDE) as a barrier to methanol crossover [21,22]. To begin the bonding procedure, a damp layer of Nafion was placed onto a Teflon sheet (10 cm × 10 cm, 0.08 mm thick). The moisture in the layer ensured temporary uniform adhesion to the Teflon sheet. Cathodic GDEs were placed catalyst side down onto the Nafion layer and a layer of aluminum foil was placed atop the microporous side of the cathodic GDE. By doing so, issues related to the GDE bonding to the surface of a Teflon sheet are avoided; although the perimeter of the Nafion membrane will bond to the aluminum foil backing, trimming with a scalpel allows easy removal of the finished cathode from the aluminum foil backing. The layers were sandwiched between two aluminum plates and then hot-pressed at 130 °C and 8 MPa. The assembly was removed from the hot-press after 5 min to yield a cathodic GDE with a Nafion membrane bonded to it.

Fuel cell assembly and testing

The microscale fuel cell assembly, shown in Fig. 2, was designed similar to a setup developed previously for catalyst and electrode characterization [23–25]. Electrodes were positioned on either side of a fuel/electrolyte flow channel, machined from polymethyl-(methylacrylate) (PMMA). The cathodic GDE was oriented to ensure the Nafion membrane was adjacent to the fuel/electrolyte channel. Copper-infused graphite plates, with catalyst access windows, served as current collectors, and gases were fed through polycarbonate flow chambers. The assembly was held together using binder clips. Catalyst deposition length (dimension parallel to the fuel inlet direction) was held constant (0.5 cm), with deposition widths of 0.5, 0.8, and 1.0 cm used in the direction normal to the fuel inlet.

A fuel stream consisting of 1 M methanol (Fisher) and 1 M sulfuric acid (GFS Chemicals) in Millipore water (18.1 MΩ) was used for all experiments. Fuel was supplied to the cell at 0.3 mL/min for catalyst deposition geometry studies and 2.0 mL/min for fuel stream temperature studies using a syringe pump (Cole Parmer). Fuel temperatures were varied between room temperature, 40 °C, 50 °C, 60 °C, and 70 °C using a nichrome resistive element wound around PTFE fuel inlet tubing and a variable current power supply. Both the heating element and the exposed fuel line are insulated with woven fiberglass to provide uniform heating of the fuel stream. The higher flow rate (2.0 mL/min) used for the fuel stream temperature studies ensured even fuel flow chamber temperature distributions by reducing heat loss to the graphite current collectors. The increased flow rate in combination with the high inlet temperatures led to elevated electrode temperatures, however forced convection was employed to maintain constant cell temperature.

Before testing, the fuel stream heater was calibrated to determine the power that must be supplied to the nichrome

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