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The effect of temperature on the output characteristics of micro direct methanol fuel cell

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HIGHLIGHTS

- A novel two-dimensional, multi-physics model is established.
- A 0.64 cm^2 metal-based μDMFC is fabricated by micro-stamping technology.
- The experimental validation with high power density is conducted.
- The experimental results are in good agreement with the simulation.

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ABSTRACT

In this paper, the effects of operating temperature on mass transport and micro direct methanol fuel cell (μDMFC) performance are presented. Furthermore, a whole two-dimensional model coupled with mass/momentum transports and temperature characteristic is established. Simulation results show that the temperature has significant effects on methanol concentration/CO_2 distributions, crossover current density, and the polarization curve. The metal-based μDMFC with the effective area of 0.64 cm^2 is fabricated using micro-stamping technology, and the detailed experimental validation is conducted. The results reveal that when the cell is supplied with a relatively low aqueous methanol flow rate, the peak power density exhibits a trend of initially going up, reaching the peak value of 85.3 mW cm^{-2} at 60 °C, and then dropping off. At the higher flow rate, however, a proportional relationship between the power density and temperature is obtained. The experimental results are in good agreement with the simulation.

1. Introduction

Conventional batteries have the disadvantage of serious environmental impact. Therefore, it is urgent to find a clean and high-energy power source for portable electronics [1–4]. Meanwhile, the micro direct methanol fuel cell (μDMFC) has been considered as a prime candidate due to the advantages of high-efficiency, low-emission, silent-operation and simplicity [5,6].

Presently, one of the most challenges for μDMFC application is the low power density. To improve the power density of μDMFC, many studies have been conducted to explore the mechanisms of methanol oxidation kinetics, methanol crossover and the mass transport inside the cell [7–11]. It is widely acknowledged that temperature has dramatic effects upon above aspects and the performance of μDMFC [12,13]. Therefore, significant attentions have been devoted to the effect of operating temperature on μDMFC performance recently. Alizadeh et al. [14] analyzed the performances of the direct methanol single cell at various cell temperatures. The results indicated that the cell performance improved with an increase of temperature in a certain range because the conductivity of the membrane and the reaction kinetics at both the anode and cathode were increased. Chen et al. [15] also investigated the effects of methanol concentration, methanol flow rate, oxygen flow rate and cell temperature on DMFC performance, and concluded that the DMFC performance increased significantly with an increase in cell temperature. In previous studies, however, only experimental investigations on the temperature effect were engaged briefly without in-depth theoretical analysis. Given the importance of temperature on cell performance, it is essential to conduct a comprehensive study on both

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simulation and experiment to fully understand the relationship between the temperature and the inner transport characteristics and cell performance.

Based on this understanding, a novel two-dimensional cell model coupled with mass/momentum transports and temperature effect was established. In this model, the methanol solution transport, CO_2 distribution and crossover current density of different operating temperatures were numerically defined. In addition, a 0.64 cm^2 stainless-based μDMFC was fabricated using micro-stamping technology, at which the effects of operating temperature on cell performance were experimentally investigated. The results from a series of experiments including polarization curve and Electrochemical Impedance Spectroscopy (EIS) showed that the μDMFC behaviors are influenced by the temperature in a complex manner. At low methanol flow rate, the power density of the cell exhibits a non-monotonic relationship with the temperature; while at higher methanol flow rate, the cell performance monotonously increased with the temperature.

2. The model analysis

A two-dimensional model was established to investigate the methanol/CO_2 transports and the temperature effect. Fig. 1 shows the calculation domain of the model. The μDMFC is assumed to be under steady-state conditions, and the diffusion layer is defined as homogeneous porous electrode.

The mass transport can be described using the Convection–Diffusion equation as follows:

$$\nabla\cdot\left(-D_{i,l}^{eff}\nabla C_{i,l}+C_{i,l}\boldsymbol{u}_l\right)=S_{i,l} \tag{1}$$

$$\nabla\cdot\left(-D_{i,g}^{eff}\nabla C_{i,g}+C_{i,g}\boldsymbol{u}_g\right)=S_{i,g} \tag{2}$$

where subscripts l/g represent liquid/gas substance,C_i is the liquid/gas concentration.D_i^{eff} is the effective diffusion coefficient, which can be modified as:

$$D_{i,l}^{eff}=D_{i,l}\varepsilon^{1.5}s^{1.5} \tag{3}$$

$$D_{i,g}^{eff}=D_{i,g}\varepsilon^{1.5}(1-s)^{1.5} \tag{4}$$

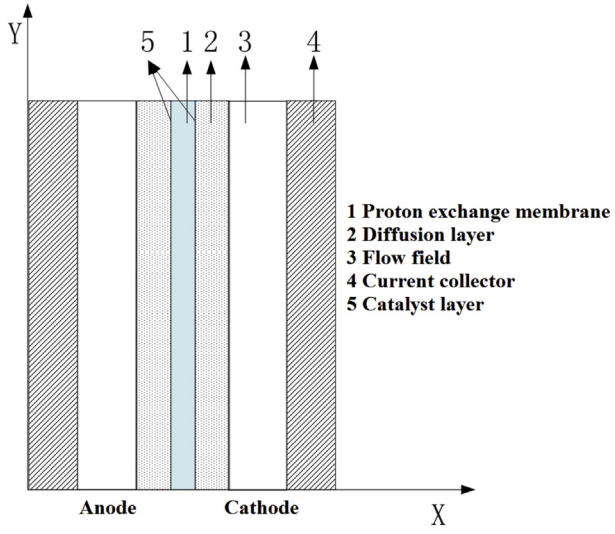

Fig. 1. Simulation domains of the model.

According to Navier–Stokes equation, the anode momentum transport suitable for methanol solution/CO_2 is given by:

$$\frac{\partial(\phi\rho_l\boldsymbol{u}_l)}{\partial t}+\nabla\cdot(\phi\rho_l\boldsymbol{u}_l\boldsymbol{u}_l)=-\nabla p_l+\nabla\cdot(\phi\mu_l\nabla\boldsymbol{u}_l)+\phi\rho_l\boldsymbol{g} \tag{5}$$

$$\frac{\partial\left((1-\phi)\rho_g\right)}{\partial t}+\nabla\cdot\left((1-\phi)\rho_g\boldsymbol{u}_g\right)=0 \tag{6}$$

where ϕ represents the liquid volume fraction.p_l is the liquid pressure in channel. $\rho$and $\boldsymbol{u}$ denote the average density and the average velocity, respectively.

The relationship between the gas and the liquid is defined as:

$$\boldsymbol{u}_g=\boldsymbol{u}_l+\boldsymbol{u}_{slip} \tag{7}$$

where,$\boldsymbol{u}_{slip}$ is the slip velocity.

Based on the ideal gas law, the relationship between CO_2 density and pressure can be described as:

$$\rho_{CO_2}=\frac{(p_l+p_{ref})M_{CO_2}}{RT} \tag{8}$$

where M_{CO_2}, R and T are molar mass, ideal gas constant and the operating temperature, respectively.

The continuity equation suitable for the porous region can be described as:

$$\nabla\cdot(\rho_l\boldsymbol{u}_l)=S_l \tag{9}$$

$$\nabla\cdot\left(\rho_g\boldsymbol{u}_g\right)=S_g \tag{10}$$

where S represents the source term.

The momentum transport in diffusion layer is defined by Darcy law:

$$\boldsymbol{u}_l=\frac{-Kk_{rl}}{\mu_l}\nabla p_l \tag{11}$$

$$\boldsymbol{u}_g=\frac{-Kk_{rg}}{\mu_g}\nabla p_g \tag{12}$$

where K and k represent the absolute permeability and the relative permeability and μ represents liquid/gas phase viscosity, k in Equations (11) and (12) can be further modified as:

$$k_{rl}=s^3 \tag{13}$$

$$k_{rg}=(1-s)^3 \tag{14}$$

where s is the liquid saturation.

Similarly, the cathode momentum transport can also be solved according to the Navier–Stokes equation:

$$\frac{\partial\left((1-\phi)\rho_g\boldsymbol{u}_g\right)}{\partial t}+\nabla\cdot\left((1-\phi)\rho_g\boldsymbol{u}_g\boldsymbol{u}_g\right)=-\nabla p_g+\nabla\cdot\left((1-\phi)\mu_g\nabla\boldsymbol{u}_g\right)+(1-\phi)\rho_g\boldsymbol{g} \tag{15}$$

$$\frac{\partial\left(\phi\rho_{H_2O}\right)}{\partial t}+\nabla\cdot\left(\phi\rho_{H_2O}\boldsymbol{u}_l\right)=0 \tag{16}$$

The oxygen diffusion and convection can be described as: