

# The Effect of Fuel and Oxidant Pumping on the Performance of a Membraneless Microfluidic Fuel Cell

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## Abstract

For the commercialization of membraneless microfluidic fuel cell, voltage and minimum power demand for various applications need a precise selection of fuel and oxidant flow rates to achieve the optimum cell performance. A numerical study on a membraneless microfluidic fuel cell was made to determine the effect of Peclet (Pe) number on the overall performance of the fuel cell. A model of 1mm (wide) x 1mm (height) x 30mm (length) Y channel configuration was validated. Formic acid was used as a fuel, oxygen was used as an oxidant and Sulphuric acid was used as an electrolyte. Creeping Flow module was selected using incompressible Navier-Stokes equations for very low Reynold number flow. Secondary Current Distribution module was used to model the electrochemical reaction and reaction rates using concentration dependent kinetics. Transport of Diluted Species module was used to account for the mass transfer between fuel and oxidant streams, in addition to species diffusivity. Pumping power increased with the increase of Pe number. The output current density slightly increased, and the net power density remained constant when Pe number was increased at Voltage greater than 0.6 V. When Pe number was increased and voltage was less than 0.6 V, the change in the output current density increased significantly, the net power density increased, and the overall efficiency dramatically decreased. The highest efficiency was achieved at the smallest Pe number. With the increase of Pe number, the change in the polarization curve, net power curve, and efficiency curve were decreased. A tradeoff must be made between the

cell efficiency and its net power output to achieve the required power output with the maximum fuel utilization.

## 1. Introduction

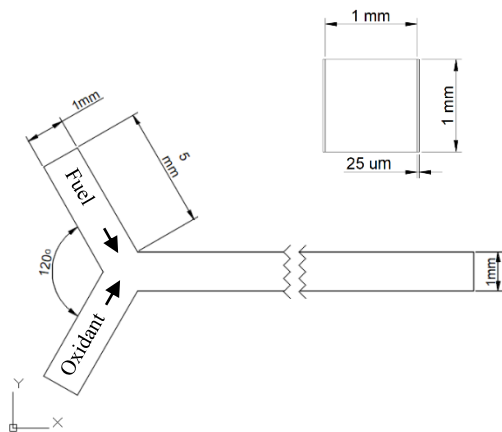
Microfluidic fuel cells is one of the most growing and promising topics that started when the need for a small, yet, high power source of energy for portable devices Emerged with an increasing demand. Studies were made on Microfluidic fuel cell with different parameters and different cases, some studies were made to improve the fuel utilization of Microfluidic fuel cells as its performance is greatly limited Due to that parameter, these studies were mainly based on the change of the geometry, or the cross section of the channel. The common shape for the channel was the rectangular cross section and the research goal was to find the effect of changing the aspect ratio of this rectangle on improving the fuel utilization. The results concluded that there was an increase in the fuel utilization at high aspect ratios [1] [2], but that was not the only parameter that affects the fuel utilization. Further research afterwards concluded that, not only high aspect ratio that governs, but also the flow rate of the fuel and oxidant that enters the micro fuel cell, the oxidant concentration, and the cathode catalyst layer thickness, each one of these parameters were probed in many parametric studies to find their effect on the fuel utilization and the performance of the micro fuel cell [3], this relation was investigated in some studies by measuring the output current density. The effect of changing a certain parameter on micro fuel cell electrochemical losses (which in turn work as performance indicator) was also studied intensively [4].

The scope of this work focuses on performing a parametric study showing the effect of changing peclot number with the change in the performance of the fuel cell.

## 2. Theory and Device Set-up

Microfluidic fuel cell have two main fluids. In this case one of them is aqueous solution of sulfuric acid saturated with oxygen as the oxidant and the other is formic acid. The two flow streams enter from separate inlets and they move beside each other as a laminar flow. The side wall of the channel of each fluid was deposited by a rectangular electrode along the length of the channel. The electrochemical reaction occurs at the interface between the fluid and the electrode results in the production of electrical power.

One of the main performance indicators of the fuel cell is the polarization curve, which is a relation between the cell output current and voltage at a certain peclot number for the same geometry and configuration. We used this polarization curve to illustrate the relation between the performance of the fuel cell and the change of peclot number, and how it will affect the net power output of the fuel cell and its efficiency. Different polarization curves were obtained at different Peclet numbers varying with a step of 1000 from 1000 to 7000.

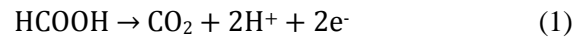


**Figure 1.** The Y-shaped Microfluidic fuel cell design with geometry dimensions and electrode assembly.

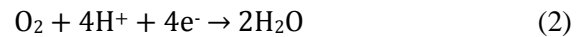
The geometry in this work is taken to be as close as possible to the geometry of Chang et.al [8] as shown in Figure. 1. The electrode – catalyst assembly was set to be 25 microns in thickness at each channel side. The length of the channel and the electrodes was set to 30 mm. on the other hand, the length of the side entry channels was set to 5mm. The width and height of both the main and entry channels was set to 1 mm. The fuel stream entered the channel from the upper side channel +Y at +30°, while the oxidant entered the channel from the lower side channel +Y at +150°.

## 3. Governing Equations

For making the simulation, it's necessary to understand the physics and equations that governs the microfluidic fuel cell performance. With the aid of the catalyst, one mole of formic acid is oxidized resulting in one mole of carbon dioxide, two protons, and two electrons.



An external circuit is used to conduct the electrons from the anode to the cathode, while the protons are transferred through the ionic conductive electrolyte, which is the Sulphuric acid.



At the cathode the electrons gathered with the protons react with the Oxygen forming water.

### 3.1 Fluid Flow

The mathematical model of the continuum fluid flow is described by the continuity equation and the Navier – Stokes equations in a 3 dimensional domain.

$$\frac{\partial \rho}{\partial t} + \nabla(\rho u) = 0 \quad (3)$$

Where the density  $\rho$  is constant for incompressible fluids.

$$\left(\rho \frac{\partial u}{\partial t} + u \nabla u\right) = \nabla P + \mu \nabla^2 u + f \quad (4)$$

Where  $P$  is the pressure,  $u$  is the velocity,  $\mu$  is the dynamic viscosity, and  $f$  is the body forces. For the case of very low Reynolds number, the creeping flow assumptions is a very good simplification of these equations. The inertia term is eliminated; resulting in the Stokes equations.

### 3.2 Charge Conservation

The electric migration of protons from anode to cathode can be represented using two modules, one of them describes the transport of charged species and the electric field within the electrolyte.

$$\sigma \nabla \cdot \nabla \phi_l = 0 \quad (5)$$

Where  $\sigma$  is the electric conductivity of the electrolyte which is constant, and  $\phi_l$  is the local potential in the electrolyte. The other module describes the voltage and current distribution within the solid phase metallic electrodes.

$$\sigma \nabla \cdot \nabla \phi_s = 0 \quad (6)$$

We can get from these equations the potentials of the solid electrode and the adjacent electrolyte to it which is needed to calculate the activation overpotential  $\eta$ .

$$\eta = \phi_s - \phi_l - E_{REVERSIBLE} \quad (7)$$

where  $E_{REVERSIBLE}$  is the reversible potential which was taken as a constant value. The calculation of the overpotential  $\eta$  is one of the parameters needed to get the current density distribution from the reaction kinetics.

### 3.3 Reaction Kinetics

The electrochemical reaction of the micro fuel cell can be represented by Butler-Volmer equation as a function of local species concentration and voltage overpotential, and it's also known as the concentration dependent

kinetics.

$$i = i_0 \left(\frac{C_i}{C_{i,ref}}\right) \left[ e^{\left(\frac{\alpha n F}{RT} \eta\right)} - e^{-\left(\frac{(1-\alpha) n F}{RT} \eta\right)} \right] \quad (8)$$

Where  $i$  is the local current density,  $i_0$  is the volumetric exchange current density of a given electrode, and this value is tuned until the simulation results match the experimental ones,  $C_{i,ref}$  is a reference concentration,  $C_i$  is the local concentration of the species "i" which refers to fuel or oxidant,  $n$  is the number of transferred electron.  $\alpha$  is the anodic and cathodic charge transfer coefficients,  $R$  is the universal gas constant,  $T$  is the operating temperature,  $F$  is the Faraday's constant and  $\eta$  is the surface overpotential. The rate of consumption of reactant species  $S$  is expressed as follows:

$$S_i = \frac{v_i i}{n F} \quad (9)$$

where  $v_i$  is the species stoichiometric coefficient.

### 3.4 Mass Transport

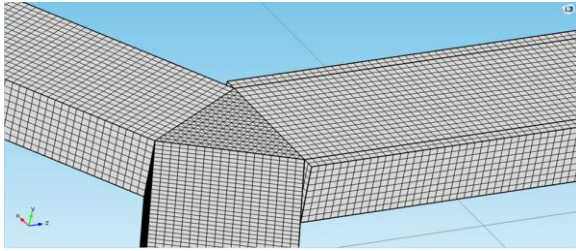
From equations (3) and (4), the values of the velocity and pressure are known, the velocity contributes to the mass transport phenomena of species. It's expected that the species is transported by convection, but due to the consumption of formic acid and oxygen, depletion region occurs, so the species at the bulk start to diffuse to the electrode wall, besides the species start to diffuse at the boundary between the fuel and the oxidant and mixing occurs at this region [7]; therefore, the species are transported by convection and diffusion. The species change in concentration is modeled using the Fick's law [9]:

$$\nabla \cdot (-D_i \nabla c_i) + u \cdot \nabla c_i = S_i \quad (10)$$

where 'i' is the reactant species,  $c$  the concentration,  $D$  is the diffusion coefficient.

#### 4. Numerical model:

In this study a 3D model of the Microfluidic fuel cell was made using Solidworks livelink feature in COMSOL to generate the geometry shown in Figure. 1. After generating the geometry, the mesh was built as shown in Figure. 2.



**Figure 2.** The Y – shaped microfluidic fuel cell model with a hybrid mesh of hexahedrons and wedges elements.

A hybrid mesh was used of a free triangular mesh on the triangle which connects both inlet and outlet channels, a swept mesh for the remaining domains was made using face meshing method “Quadrilateral” (Generate hexahedrons) with a finer mesh size setup. The resulting average mesh quality was 0.9966 with total number of elements 136,890, with this results the mesh was quite acceptable.

Creeping flow model was added to the physics to account for the effect of low Reynolds number as this model neglects the inertial terms from Navier-Stokes equations leading to Stokes equations. Secondary current distribution model was used to add the governing equation concerning the conservation of charges in the electrode and electrolyte. Concentration dependent kinetics model was used instead of the ordinary butler – Volmer model as the latter doesn’t account the concentration of the species and its effect in the local current density. Transport of diluted species model was used to implement the mass transport equation in the study. Coupled-stationary step solver was used in the study to solve the three models simultaneously.

Parametric swept study was made to change the peclot number from 1000 to 7000 with step of 1000, for each peclot number, another parametric

study was made for the polarization curve between cell voltage and local current density, it started from 0.2 v to 0.8 v with step of 0.1 v. The swept type of all combinations was used to get the polarization curve at each peclot number.

A workstation computer with 126 GB memory and 30 cores was used for the validation that took 32 hours in duration, while the parametric study took almost 120 hours.

#### 5. Simulation Results

A validation for the model was done by using the data of Chang et al. [8]. The results were found to be reasonable with a minor error at cell voltage of 0.2 v due to the high gradient of the concentration at the boundary of the cell as a result of increasing consumption of species.

Polarization curves and net power density of different peclot numbers for the same channel geometry were studied. For the same Cell voltage, as shown in Figures. 3, and 4, it’s noticed that the increase in peclot number is accompanied by an increase in the local current density and the power density as well. This happened Due to a couple of reasons. First, the depletion region decreased by increasing the Fuel and Oxidant flow rate, replacing the consumed flow at the electrode surface in a shorter time, So that the overall chemical reaction process is enhanced. Second, the diffusion region at the boundary between the fuel and the oxidant decreased, according to the scaling law [5] for pressure driven laminar flow of two aqueous solutions:

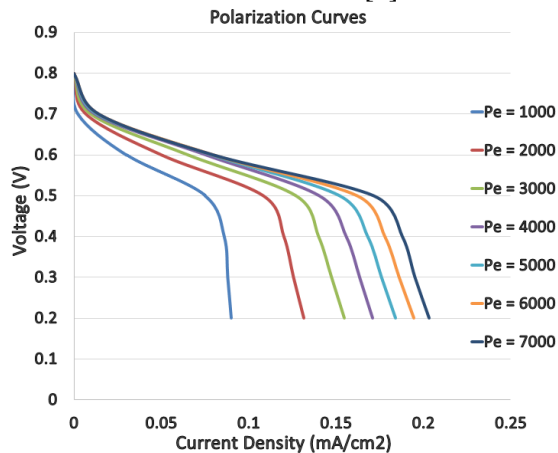
$$\delta \propto \left(\frac{DH\alpha}{U}\right)^{\frac{1}{3}} \quad (11)$$

Where  $\delta$  is the width of the diffusion region between fuel and oxidant, D is the diffusion coefficient, U is the flow velocity, z is the downstream position and H is the channel height. Fluid crossover was limited as the flow speed increases, and therefore the power density and the overall performance of the cell were increased.

Due to increasing peclot number the gradient in the output current density decreased. for a change in peclot number from pe = 1000 to pe = 2000, the current density jumped from 0.086 (mA/cm<sup>2</sup>) to 0.12 (mA/cm<sup>2</sup>) with a difference of

(0.34) (mA/cm<sup>2</sup>), and the net power density increased by 16.5 (μw/cm<sup>2</sup>), on the other hand when peclot number changed from pe = 6000 to pe = 7000, the current density increased slightly by 0.01 (mA/cm<sup>2</sup>) and the net power density increased only by 4.6 (μw/cm<sup>2</sup>).

It could be assumed that at very high peclot number the polarization curve and the net power density will not change, which is due to the dramatic decrease in depletion region along the electrodes and the diffusion region between the fuel and the oxidant which are assumed to be eliminated, so that the cell will obtain its maximum current density and net power output. However, this won't be practical, as the increase in peclot number is limited by the laminar flow assumption, when peclot number exceeds certain limits, hydrodynamic instability may occur and the results will not be accurate [6].



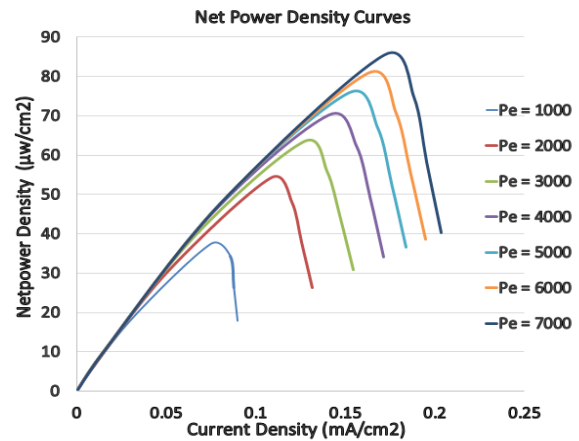
**Figure 3.** Polarization curves with current density normalized by electrode surface area for different peclot number values

For the efficiency shown in Figure. 5, it was anticipated that the efficiency decreased with the increase in peclot number, and that's because of the large amount of unconsumed fuel that came out of the cell without contributing into the reaction. The efficiency is calculated as follows:

$$\eta = \frac{IV}{PQ + m \cdot f \cdot C.V} \quad (12)$$

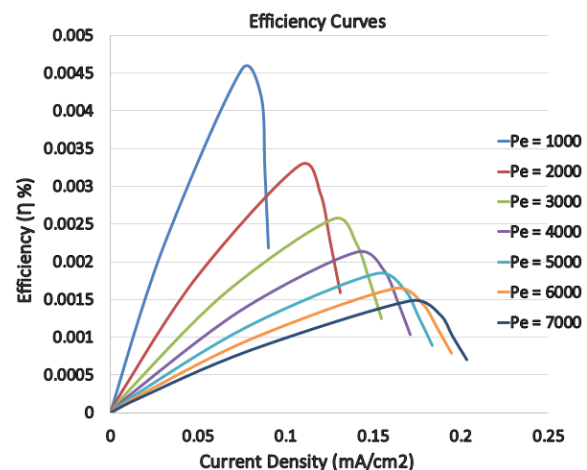
Where I is the current density, V is the cell voltage, P is the pressure at inlet, Q is the flow

rate of both fuel and oxidant, m·f is the fuel flow rate, and C.V is the fuel calorific value (C.V = 13000 kJ/kg).



**Figure 4.** Net power density curves versus current density normalized by electrode surface area for different peclot number values

The pumping power didn't contribute a lot in the overall cell performance, the pumping pressure increased linearly by only 3.2 Pascal when peclot number was increased by 1000. Therefore the dependency of the efficiency on the fuel flow rate is greater than its dependency on the cell pumping power.



**Figure 5.** Efficiency curves versus current density normalized by electrode surface area for different peclot number values.

## 6. Conclusions

A Multiphysics model of a Y channel configuration was validated using data from the literature. The effect of Peclet (Pe) number on the overall performance of the fuel cell was demonstrated using a parametric study of various values of it. The results showed that effect of the increase in pumping power with the increase in Pe number was linear and negligible compared to the increase in the net power density. While the efficiency and fuel utilization are heavily affected by the increase in Pe number. Also the effect of the change in Pe number on the performance decreases as its value increases. Future work should address the modifications over the geometry under various operating conditions and requirements, and the control of fuel and oxidant flow rates, with the integration of micro Pumps and Storage units to meter the flow for maximum working hours of the device.

## 7. References

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## 8. Acknowledgements

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## 9. Appendix

Parameter	Anodic flow	Cathodic flow
Density, $\rho$ (Kg m <sup>-3</sup> )	1000	
Dynamic viscosity, $\mu$ (Kg m <sup>-1</sup> s <sup>-1</sup> )	0.001	
Diffusion coefficient, D	8.3 x 10 <sup>-10</sup>	
Inlet concentration, C (mol m <sup>-3</sup> )	2100	0.5
Ionic conductivity, $\sigma$ (S m <sup>-1</sup> )	11.47	43
Exchange current density, $i_0$ (A m <sup>-3</sup> )	3.82 x 10 <sup>5</sup>	100
Charge transfer coefficient, $\alpha$	0.5	0.5
Number of electrons transferred, n	2	4
Reversible potential, E <sub>REVERSIBLE</sub> (V)	0	0.9
Reference concentration C <sub>ref</sub> (mol m <sup>-3</sup> )	2100	0.5
Faraday constant, F (C mol <sup>-1</sup> )	96485.34	
Cell temperature, T (K)	298	
Universal gas constant, R (J mol <sup>-1</sup> k <sup>-1</sup> )	8.314	