MODELING AND SIMULATION OF METHANOL CROSSOVER EFFECT IN THE DIRECT METHANOL FUEL CELL (DMFC)

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CHEMICAL ENGINEERING

UNIVERSITI TEKNOLOGI PETRONAS

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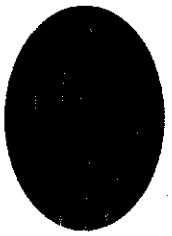

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by

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10722

Dissertation submitted in partial fulfilment of
the requirements for the
Bachelor of Engineering (Hons)
Chemical Engineering

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CERTIFICATION OF APPROVAL

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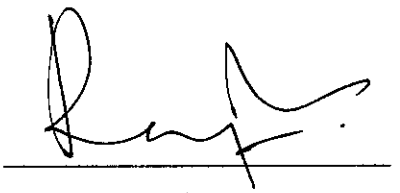
Approved by,



(Dr. Rajashekhar Pendyala)

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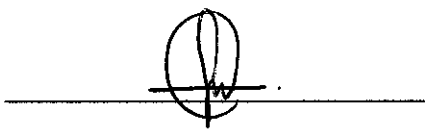
SEPTEMBER 2011

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.



(Mohamad Syazwan Bin Ahmad Zubir)

ABSTRACT

The interest in direct methanol fuel cell (DMFC) is based on their potential for energy saving and cleaner energy production. However, there are some limitations that DMFC has to overcome before it can become one of the main alternative power sources in the market.

This project will focused on the methanol crossover effect as one of the limitation that DMFC have. CFD model was developed using Ansys Fluent PEM Fuel Cell Module to study and understand the behaviour of the effect. Temperature will be the main varying parameters that will be studied for this project.

Simulation results show that methanol crossover significantly increases the cathode overpotential at low current density but it is reduced when the current density is increased to a certain amount of value.

ACKNOWLEDGEMENT

This dissertation would not have been possible without the guidance and the help of several individuals who in one way or another contributed and extended their valuable assistance in the preparation and completion of this study.

First and foremost I offer my sincerest gratitude to my supervisor, Dr. Rajashekhar Pendyala, who has supported me throughout this project with his patience and knowledge whilst allowing me the room to work in my own way.

Not forgetting thanks to Final Year Project II coordinator, Dr. Lukman b Ismail and all Chemical Engineering Department lecturers for the help and beneficial professional guidance to me.

Last but not least, thank you to my family for their love, support and encouragement.

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LIST OF ABBREVIATIONS

CFD	Computational Fluid Dynamics
DMFC	Direct Methanol Fuel Cell
PEM	Polymer Electrolyte Membrane
CO_2	Carbon Dioxide
CH_3OH	Methanol
H_2O	Water
O_2	Oxygen
Pt	Platinum
Ru	Ruthenium
V-I	Voltage-Current
UDF	User-Defined Function
UDS	User-Defined Scalar

CHAPTER 1
INTRODUCTION

1.1 Background Study

Petroleum, the world's most productive fuel, has given rise to several environmental problems. The burning of fuels derived from petroleum produces emissions which carry much of the responsibility for air pollution and greenhouse gas effect. In order to reverse the trend of destroying the nature, a change to a more environmentally friendly resource of producing energy is desirable. Fuel cell is one of the best options. It need no particular environment to work well and is highly efficient both electrically and physically. Its principle is the reverse of electrolysis process where fuel cell generates electricity by combining oxygen and hydrogen molecules instead of separating them from water.

Fuel cells can be divided into several types based on the electrolytes used in it. Direct Methanol Fuel Cell (DMFC) is one of them. Operated at relatively low temperature and creating little heat with the operating temperature around 50°C to 120°C, DMFC is considered the best fuel cell type for mobile devices because of its compact size. It can works directly on methanol without having to first convert those fuels into hydrogen gas. Methanol is an attractive liquid fuel because it is relatively cheap, easily stored and handled, readily available and soluble in aqueous electrolytes, and has a high energy density. (Viswanathan & Scibioh, 2007)

DMFC consists of an anode at which methanol is electro-oxidised to CO_2 through the reaction,

$$CH_3OH + H_2O \rightarrow CO_2 + 6H^+ + 6e^-$$

And a cathode at which oxygen (usually as air) is reduced to form water or steam,

$$^3/_2\, O_2 + 6e^- + 6H^+ \rightarrow 3H_2O$$

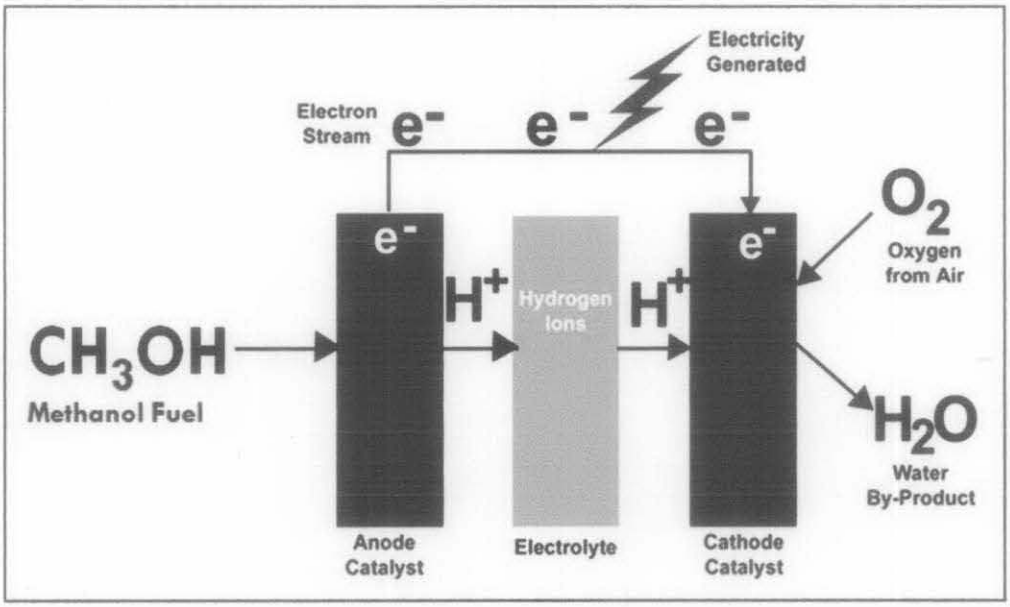


Figure 1: Schematic diagram of direct methanol fuel cell operation

In order to make it possible for DMFC to be applied to the real energy consumers such as portable electronics and automotive industry, polymer electrolyte membrane or PEM is applied. This PEM technology is an important part in DMFC because of its attribute where it can operate a fuel cell with a low pressure and temperature operation and no liquid electrolyte. Nafion® perfluorosulfonic acid polymers are the commonly used fuel cell membranes.

1.2 Problem Statement

DMFC technology is a further development of polymer electrolyte membrane fuel cell (PEMFC). It has given the advantage to the polymer electrolyte membrane technology where the fuel which is aqueous methanol, does not need to be converted to hydrogen first. The methanol fuel can be directly use in the cell as a high energy density liquid fuel.

However, the fuel itself is also the cause of its main drawback which is lower efficiency and lower power density compared to PEMFC. Although it would be desirable that methanol could be spontaneously oxidised at anode, a methanol transport across the membrane, has been observed. It causes depolarization losses at the cathode and conversion losses. In order to improve the performance of the DMFC, it is necessary to eliminate or at least reduce the loss of fuel across the cell, usually termed methanol crossover. (Heinzel & Barragán, 1999)

1.3 Objectives

The objectives of this research project are:

1. To model and simulate single channel direct methanol fuel cell (DMFC) by using Computational Fluid Dynamics.
2. To study and further understand the effect of methanol crossover in DMFC performance under the influence of temperature.

1.4 Scope of Study

The purpose of this project is to understand the effect of methanol crossover in the performance of direct methanol fuel cell that uses polymer electrolyte membrane as its electrolyte. It will take into account various possible operating conditions of DMFC such as thickness of the membrane, operating temperature and pressure, and concentration of methanol fuel.

This research will make use the knowledge of computational fluid dynamics (CFD) to understand the behaviour of the fuel cell electrochemistry. This will be done by using ANSYS Workbench as well as ANSYS Fluent simulation software. In Fluent, there is an add-on module called the Fuel Cell User Defined Function (UDF) that helps researchers to analyse and optimize fuel cell design as well as performance. (Ansys, 2011)

Throughout the research, UDF will be used to model and simulate electrochemical reactions, potential field boundary conditions, current density distribution and cell voltage. Other transport processes such as fluid flow, heat transfer and mass transfer will be considered as well to simulate the methanol cross over behaviour across the membrane.

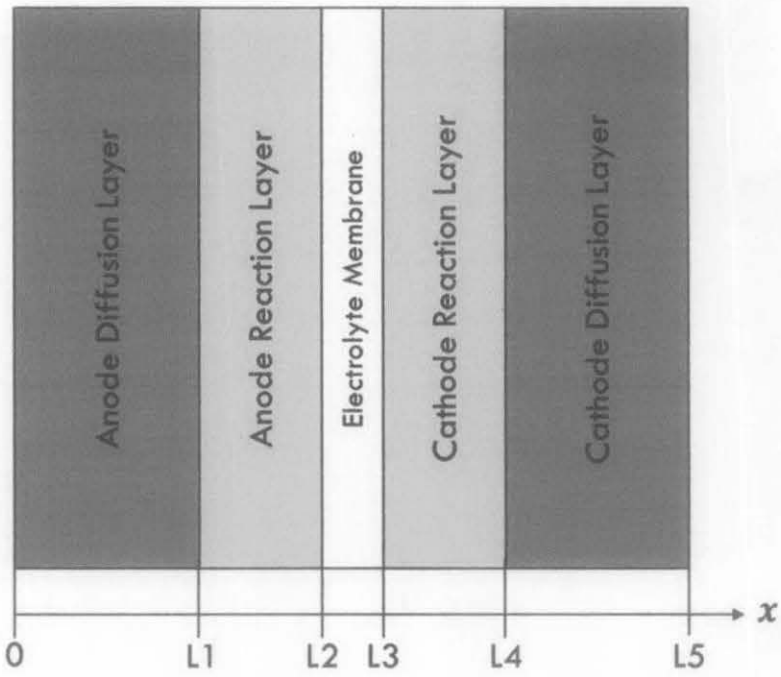


Figure 2: Proposed domain and geometry for direct methanol fuel cell simulation

Figure 2 shows the proposed domain by for the DMFC model that will be used for the simulation later (Zhang & Wang, 2004). It contains 3 main parts which are:

- Diffusion layers (anode and cathode)
- Reaction / Catalyst layers (anode and cathode)
- Polymer electrolyte membrane

After modeling and simulation, the final part of this research will be the validation of the result by comparing it with an experimental data from related journal. The model will be revised and modified according to the real data before the conclusion and recommendation is made.

CHAPTER 2
LITERATURE REVIEW

2.1 Methanol Crossover

Methanol cross-over can be defined as the permeation of methanol through the electrolyte membrane. Polymer electrolyte membrane is not fully impermeable to methanol and allow for significant quantities to permeate from the anode to the cathode. Two key mechanisms are identified which are:

- Diffusion
- Electro-osmotic drag

When methanol permeates through the polymer membrane and reaches the cathode, it will oxidized and leads to a mixed potential and decrease in cell voltage. Furthermore, the oxidized methanol is effectively wasted fuel with clear negative impact in the overall efficiency of the cell which further reduces the energy yield of the cell.

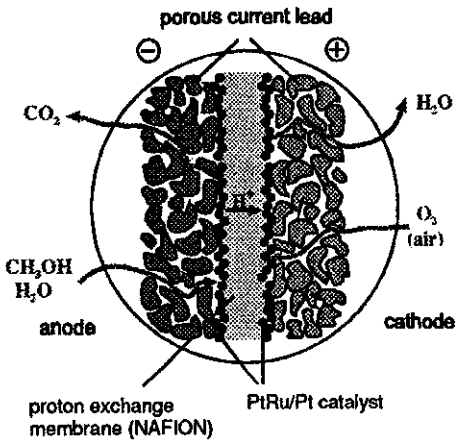


Figure 3: Model of DMFC structure

The basic scheme of a DMFC fuel cell is shown in Figure 3. The methanol / water reaction mixture diffuses through the porous anode to the electrochemically active reaction zone in the immediate area of the membrane, where the electrochemical oxidation of the methanol takes place. The CO_2 reaction product diffuses back into the anode channels.

The protons formed entrain certain amounts of water into the solvating envelope and under the influence of the electric field, migrate to the cathode where they react with oxygen to form water after electron acceptance. In addition, an undesired methanol transport from the anode to the cathode takes place caused by diffusion and electro osmotic effects. The methanol fractions arriving at the cathode react with the oxygen present there to form CO_2.

Much effort has been made to eliminate or at least to reduce the methanol crossover effect to some degree (Song, et al., 2004) which includes:

- Adopting high effective anode catalysts and methanol-tolerant cathode catalysts
- Optimizing the feed concentration of methanol aqueous solutions
- Developing novel electrolytes or doping Nafion® membranes

Many researches around the world have recently studied about methanol cross-over effect in DMFC both experimentally and theoretically. Dohle et al. and Valdez and Narayanan et al. measured the methanol cross-over flux with different membrane thickness and showed that the methanol cross-over rate is inversely proportional to membrane thickness, hence indicating that diffusion is dominant.

In addition, Ren et al. compared diffusion with electro-osmotic drag processes and demonstrated the importance of electro-osmotic drag in methanol transport through the membrane. Valdez and Narayanan and Dohle et al. studied the temperature effects on methanol cross-over and showed that the methanol cross-over rate increases with cell temperature. Wang et al. analysed the chemical compositions of the cathode effluent of a DMFC with a mass spectrometer. They found that the methanol crossing over the membrane is completely oxidized to CO_2 at the cathode in the presence of Pt catalyst (Casalegno & Marchesi, 2008).

2.2 Experimental Data

The following figures show the experimental result taken from several journals regarding the methanol cross-over in DMFC. These data will be used later on to validate the result gain from the CFD modeling and simulation.

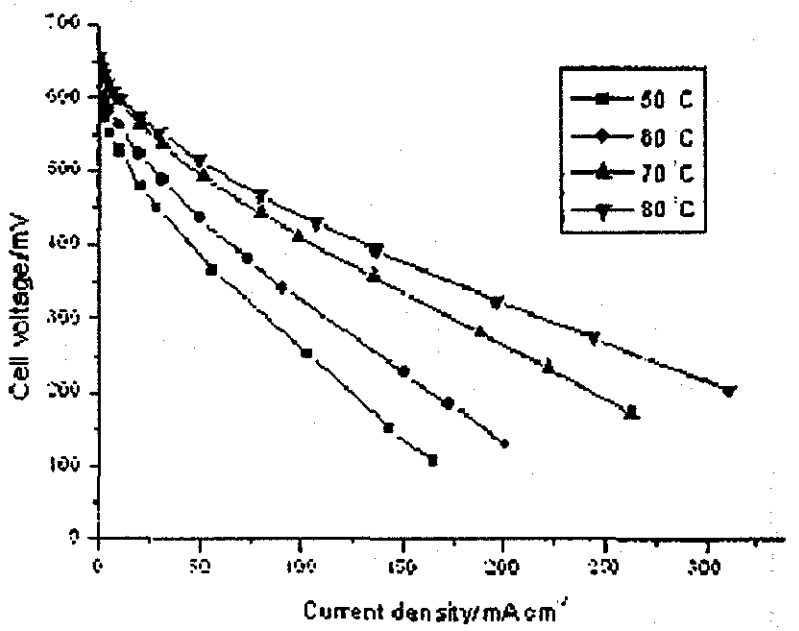


Figure 4: Effect of operating temperature on DMFC V-I curve (Zhang & Wang, 2004)

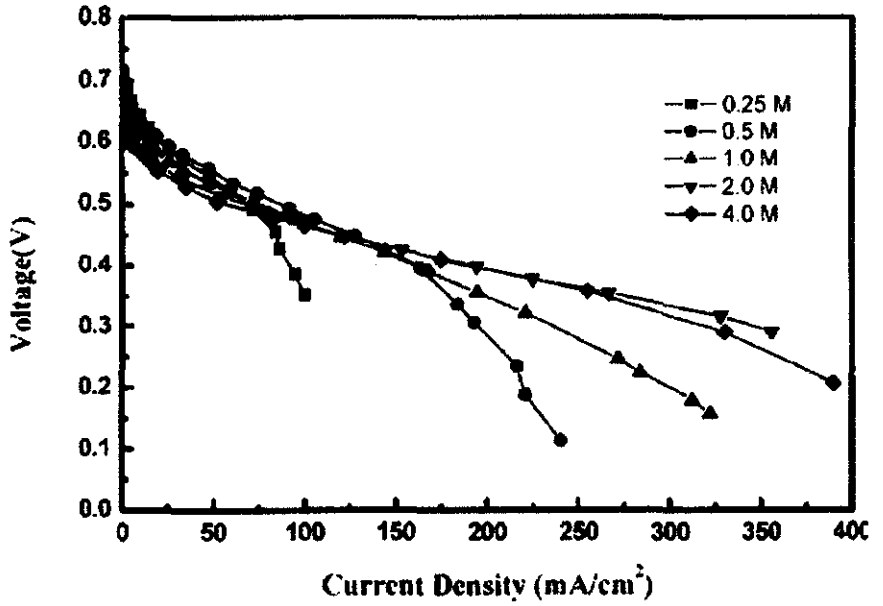


Figure 5: Effect of methanol concentration on DMFC performance (Song, et al., 2004)

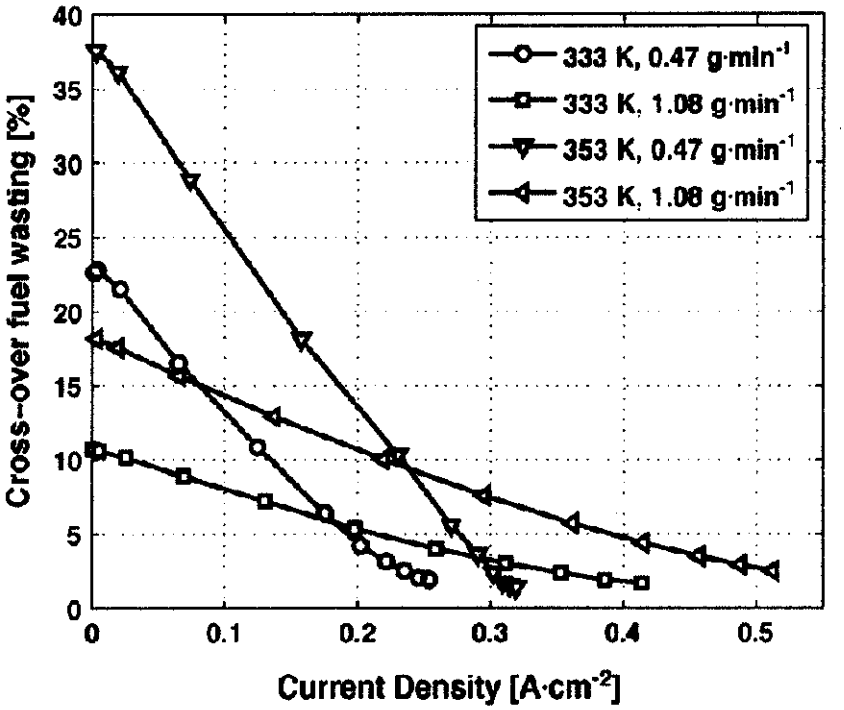


Figure 6: Cross-over fuel wasting (Casalegno & Marchesi, 2008)

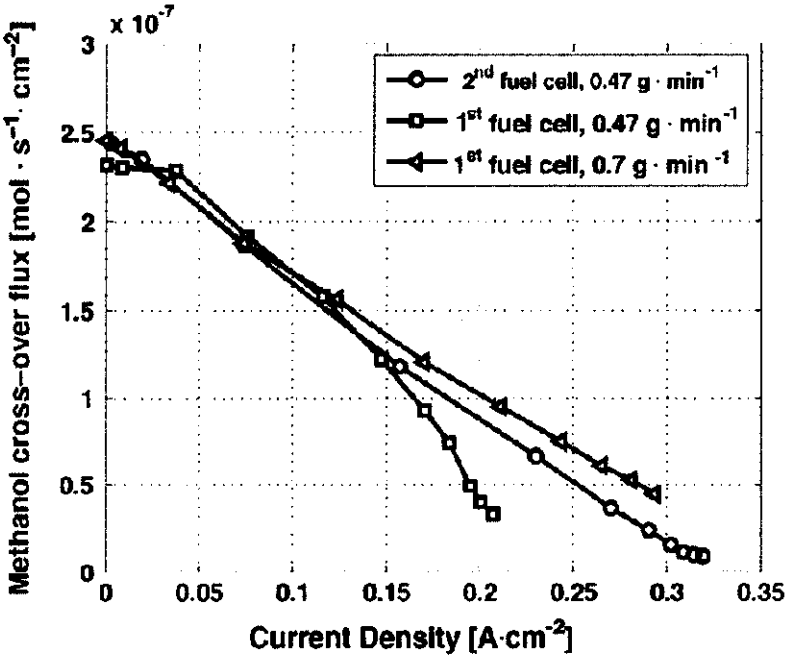


Figure 7: Comparison of methanol cross-over flux (Casalegno & Marchesi, 2008)

2.3 Modeling Equations

Figure below represent division of the DMFC into functional regions. The mathematical modeling equation will be based on the respected process (Dohle, Divisek, & Jung, 2000).

Table 1: Division of the cell into functional region

Diffusion Layer	• Diffusion • Permeation in the gas phase
Catalyst Layer	• Diffusion • Electro-osmosis • Reaction
Membrane	• Diffusion • Electro-osmosis
Catalyst Layer	• Diffusion • Electro-osmosis • Reaction
Diffusion Layer	• Diffusion • Permeation in the gas phase

2.3.1 Diffusion Layer

Transport in the gas diffusion layer is modelled as transport in porous media. The diffusion takes place in the transition region by a superposition of Knudsen diffusion and Stefan–Maxwell diffusion:

$$\frac{N_i^d}{D_i^k} + \sum_{j=1}^{n} \frac{y_j N_i^d - y_i N_j^d}{D_{i,j}^m} = -c_{tot} \frac{d_{y_i}}{d_x}$$

Molar flows due to a total pressure gradient are taken into consideration by the Darcy expression:

$$N_i^p = -y_i B_i \frac{d_{c_{tot}}}{d_x}$$

The electronic current produced or consumed in the catalyst layers flows through the gas diffusers towards the channel structure. This electronic current leads to a voltage drop via Ohm's law:

$$i_{el} = \sigma_{el}^{eff} \nabla_{\varphi_{el}}$$

2.3.2 Catalyst Layer

Catalyst layer is modelled as a porous body which is filled with ionically conducting material. The local current density distribution in the catalyst layers for the cathodic reaction is modelled based on Butler-Volmer equation:

$$i_c = i_{O,ref}^c \frac{P_{O_2}}{P_{O_{2ref}}} exp[-\frac{\alpha_c F}{RT}(\varphi_{el} - \varphi_{ion} - \varphi_{O,ref}^c)]$$

While for the anodic reaction:

$$i_a = i_{O,ref}^a \frac{P_{MeOH}}{P_{MeOH,ref}} exp[+\frac{\alpha_a F}{RT}(\varphi_{el} - \varphi_{ion} - \varphi_{O,ref}^a)]$$

The source-sink terms of the respective material are linked via Faraday's law to the respective local current density:

$$QS_{Methanol} = -\frac{i_a}{6F}$$

2.3.3 Membrane Electrolyte

The balance between the electro-osmotic drag of water from anode to cathode and back diffusion from cathode to anode yields net water flux through the membrane:

$$N_w = n_d M_{H_2O} \frac{i}{F} - \nabla.(\rho D_w \nabla y_w)$$

The water diffusivity in the polymer can be calculated as follow:

$$D_w = 1.3 \times 10^{-10} exp[2416\left(\frac{1}{303} - \frac{1}{T}\right)]$$

CHAPTER 3
METHODOLOGY

3.1 Computational Fluid Dynamics

Computational Fluid Dynamics (CFD) is the analysis of systems involving fluid flow, heat transfer and associated phenomena such as chemical reactions by simulation.

An electrochemical model will be build using GAMBIT and FLUENT software, suitable to the chemical reactions for DMFC and PEMFC, which are used to predict local current density and voltage distributions at electrolyte surfaces. Gambit is a pre-processor tool for FLUENT to setup geometry and generate mesh. The geometry is then export to FLUENT where it runs the simulation corresponding to the geometry. (Ansys, 2011)

This project will make use of the PEM Fuel Cell Model that has been integrated in the FLUENT software. The Model is comprised of several of several user defined functions (UDFs) and a graphical user interface. The potential fields are solved as user-defined scalars. The electrochemical reactions occurring on the catalyst are modeled through various source terms while other model parameters are handled through the user interface.

For the purpose of validating the result, WinDIG software is used. WinDIG is a free data digitizer which is very useful to get back curves and points from scanned images. This is an important process as the value and graph obtained for the experimental result is not in digital form.

3.2 Software Required

- Ansys Fluent
- Gambit
- WinDIG
- Microsoft Office

3.3 Project Flow Chart

Figure below shows the steps taken in doing the CFD simulation.

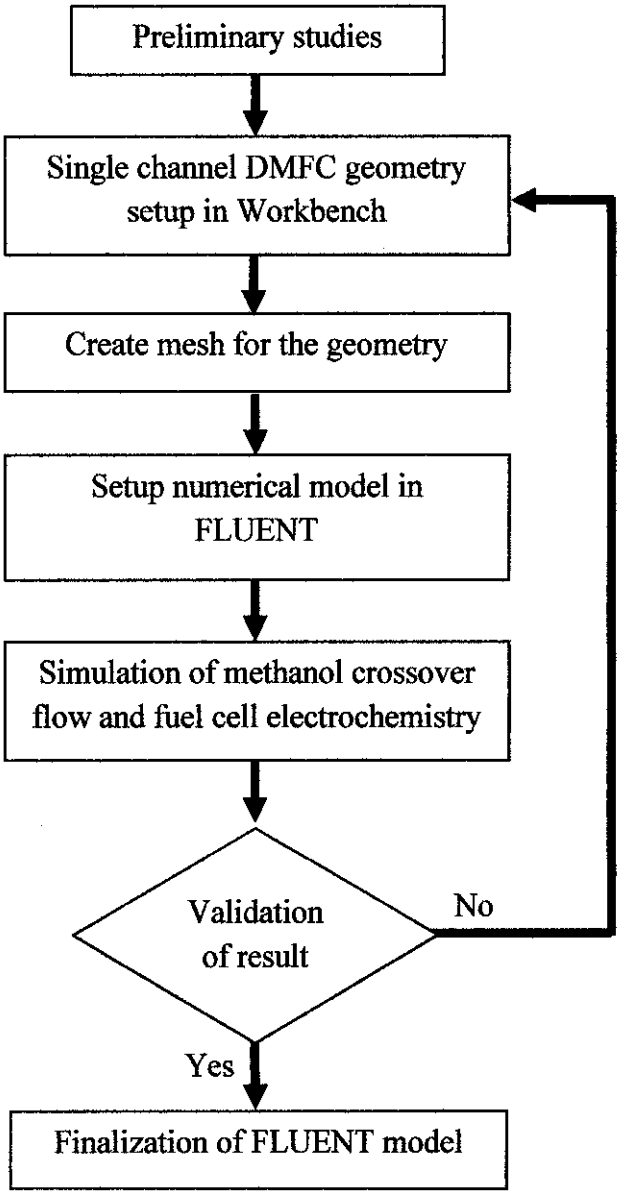


Figure 8: Project flow chart

CHAPTER 4
RESULT AND DISCUSSION

4.1 Geometry and Mesh

Below are the diagrams for geometry of DMFC created using Gambit software. At this stage, the grid of the model is defined as well by creating the mesh to the geometry created earlier. The meshing is important as it will be used later for calculation of the numerical model on each individual node.

Table below shows the dimension used in constructing the geometry and mesh (Yang & Zhao, 2009):

Table 2: Geometrical parameters

Name	Dimension (mm)
Diffusion layer	0.26
Catalyst layer	0.02
Membrane	0.13
Channel width, x	1.0
Channel height, y	1.2
Channel length, z	50

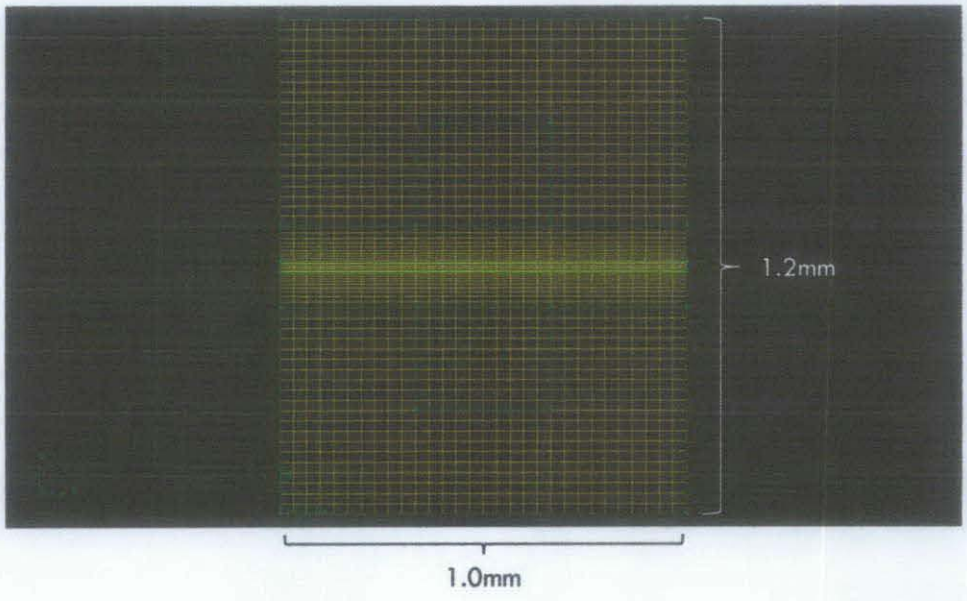


Figure 9: Front view of the DMFC geometry and mesh

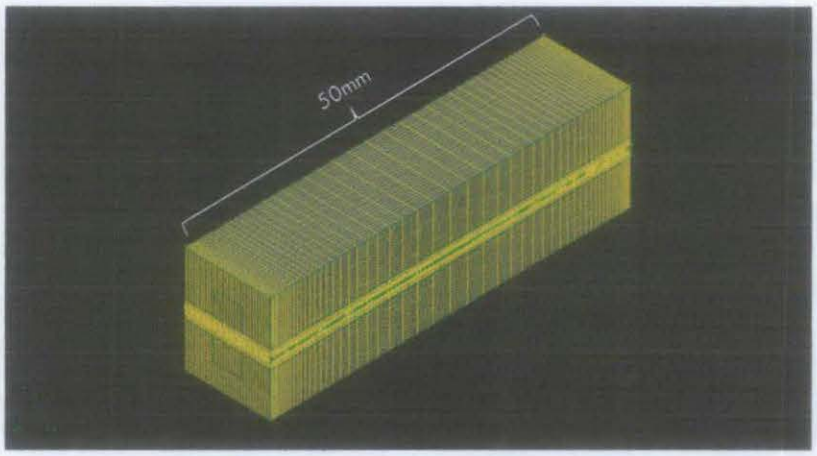


Figure 10: Isometric view of the DMFC geometry and mesh

4.2 Cell and Boundary Zones

The following regions are defined in the fuel cell mesh in order to simulate it using Fluent Fuel Cell Model:

Table 3: Cell zones

Anode	Cathode
Current Collector	Current Collector
Flow Channel	Flow Channel
Diffusion Layer	Diffusion Layer
Catalyst	Catalyst
Electrolyte Membrane	

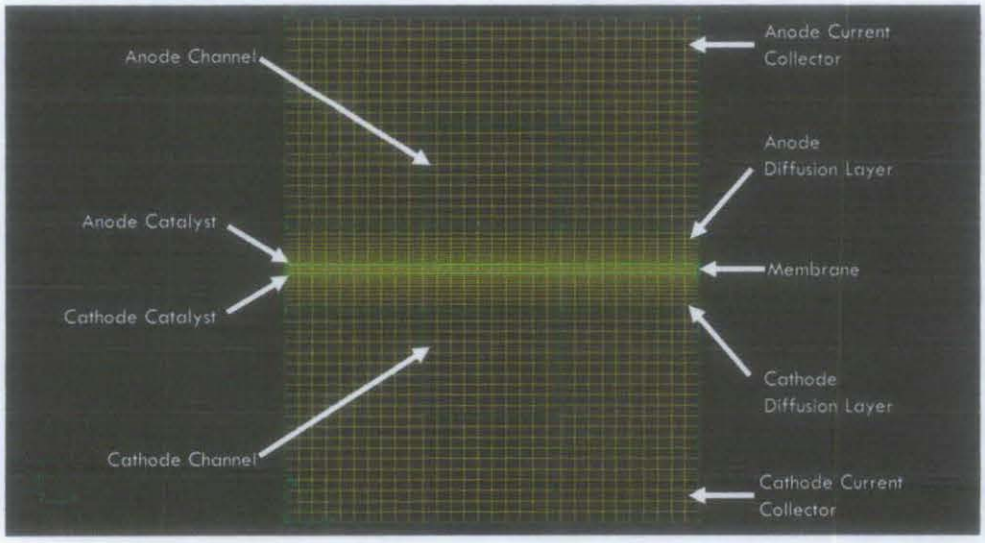


Figure 11: Cell zones

Table 4: Boundary zones

Anode	Cathode
Inlet Channel	Inlet Channel
Outlet Channel	Outlet Channel
Terminal	Terminal

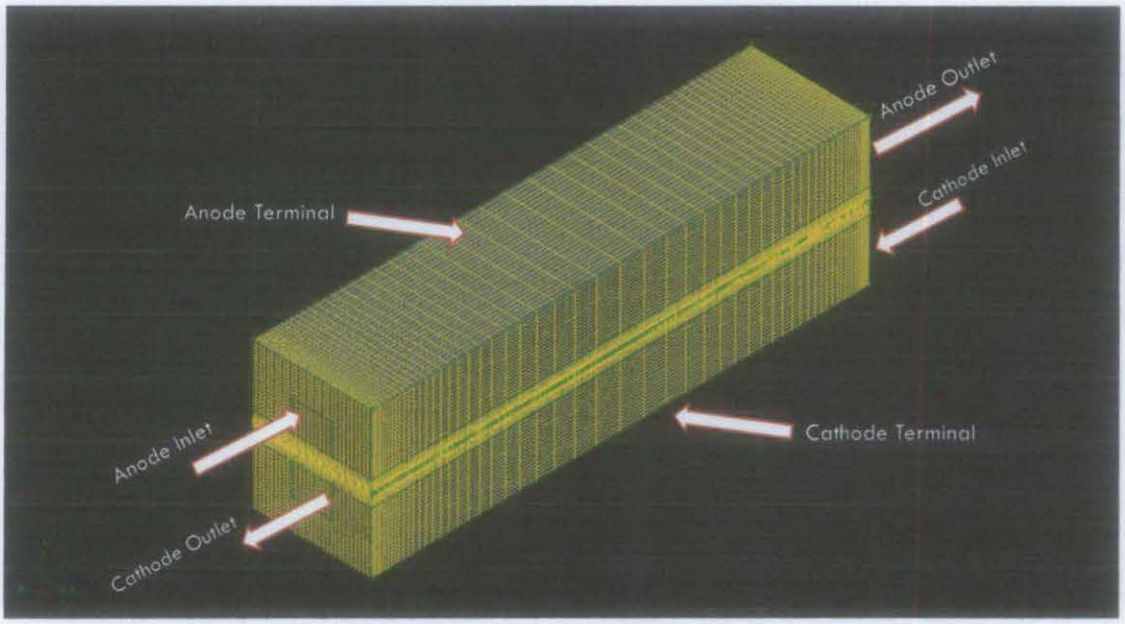


Figure 12: Boundary zones

4.3 Simulation Results

In this simulation, the varying parameter that will be focused is the temperature value. The temperature will be varies from 60°C, 70°C and 80°C. Current-density and voltage of the fuel cell will be recorded for each corresponding temperature.

Table 5: Cell operating parameters

Parameters	Value
Operation Temperature	60°C, 70°C, 80°C
Anode / Cathode Inlet Pressure	0.1MPa
Methanol Concentration	2.0 mol L^{-1}

The results shown below are corresponding to 750mV cell voltage. Other results can be obtained in appendix B and C.

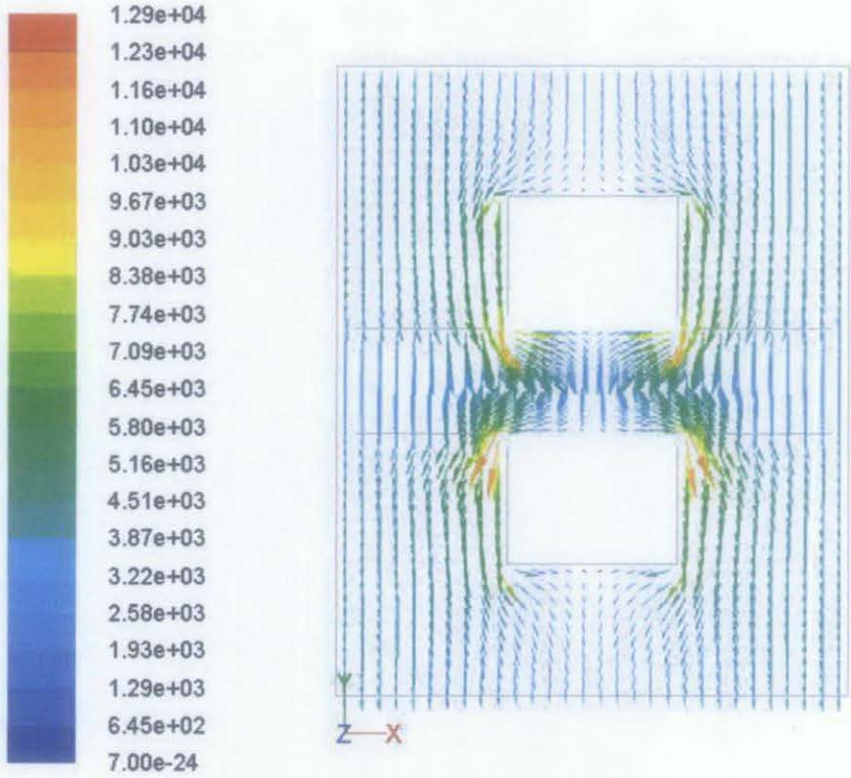


Figure 13: Current flux density distribution at 60°C

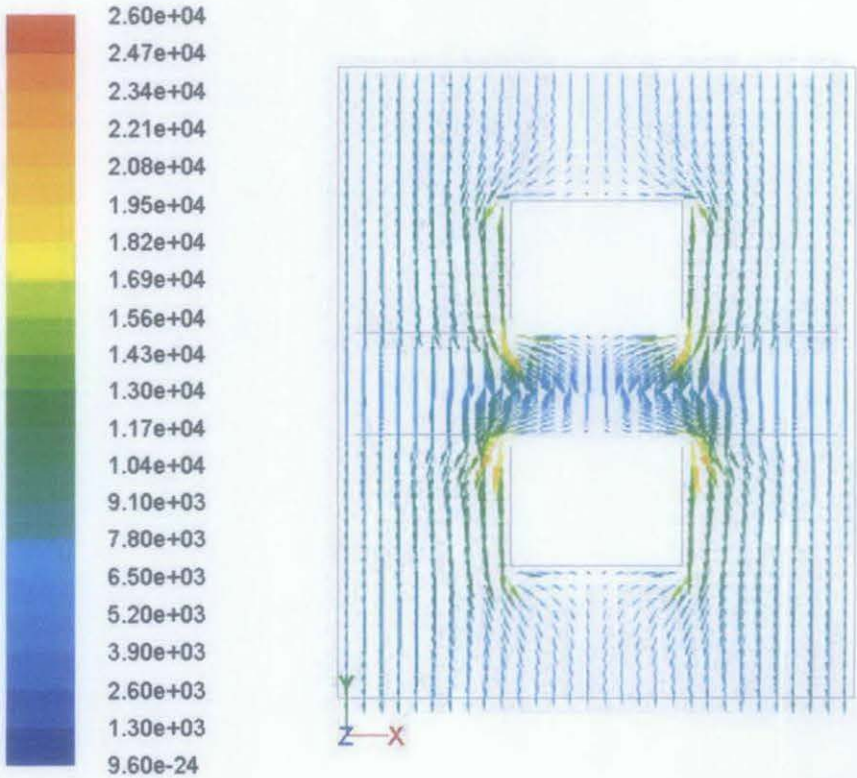


Figure 14: Current flux density distribution at 70°C

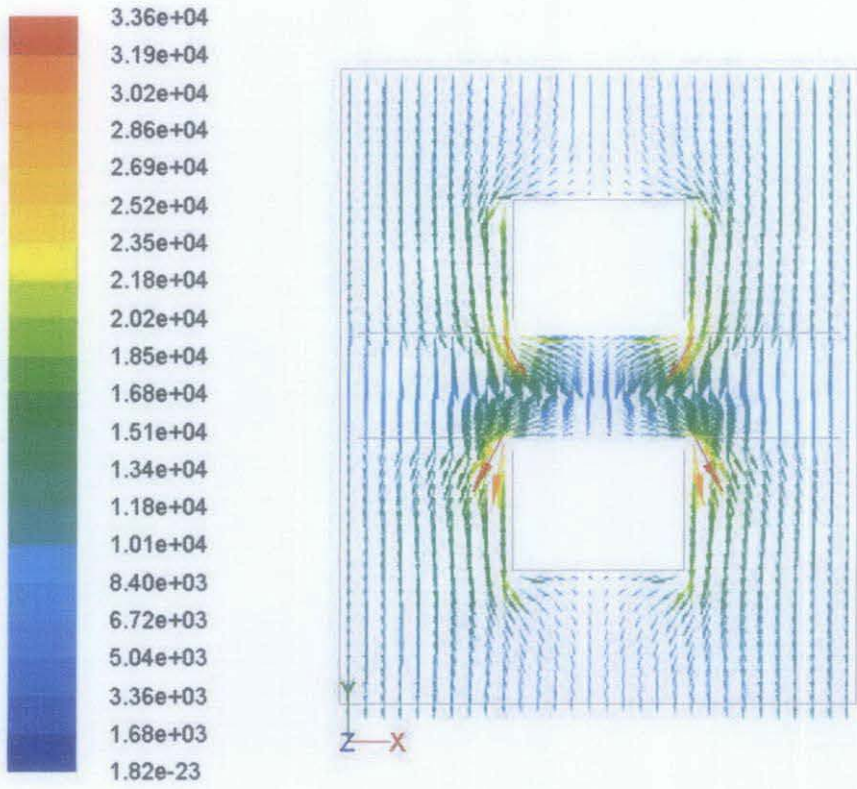


Figure 15: Current flux density distribution at 80°C

The overall simulation results for 60°C, 70°C, and 80°C is plotted in the voltage-current curves as shown below:

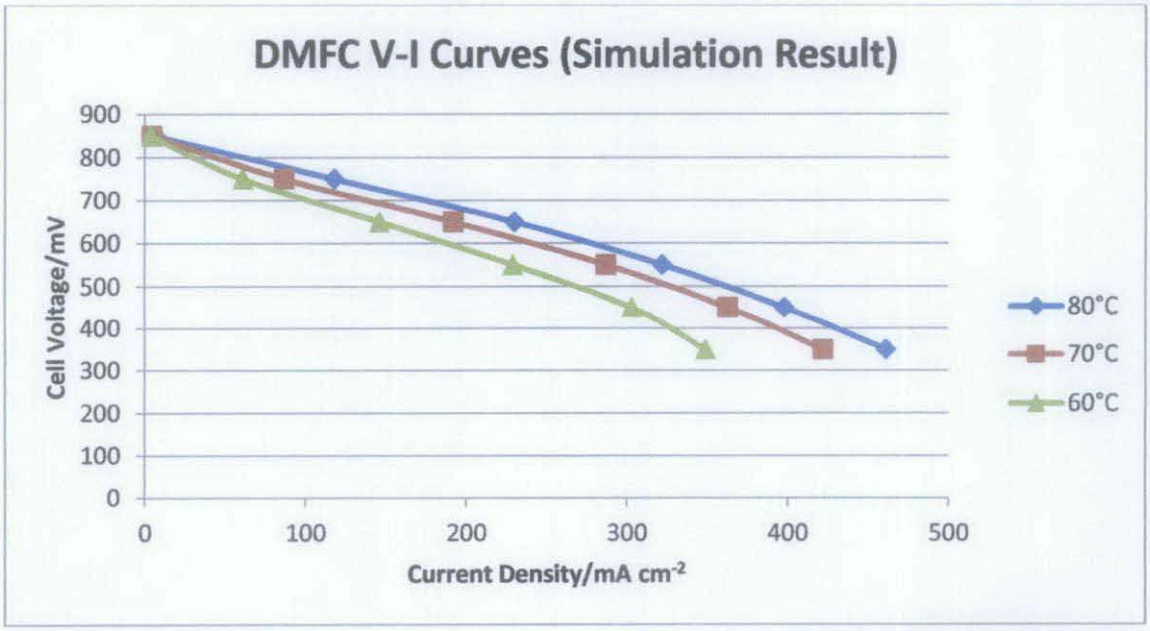


Figure 16: Effect of operating temperature on DMFC V-I curve (simulation result)

4.4 Data Validation

The simulation results are validated by comparing it with an experimental data (Zhang & Wang, 2004). The data are extracted by using WinDIG software and it is shown in graph below:

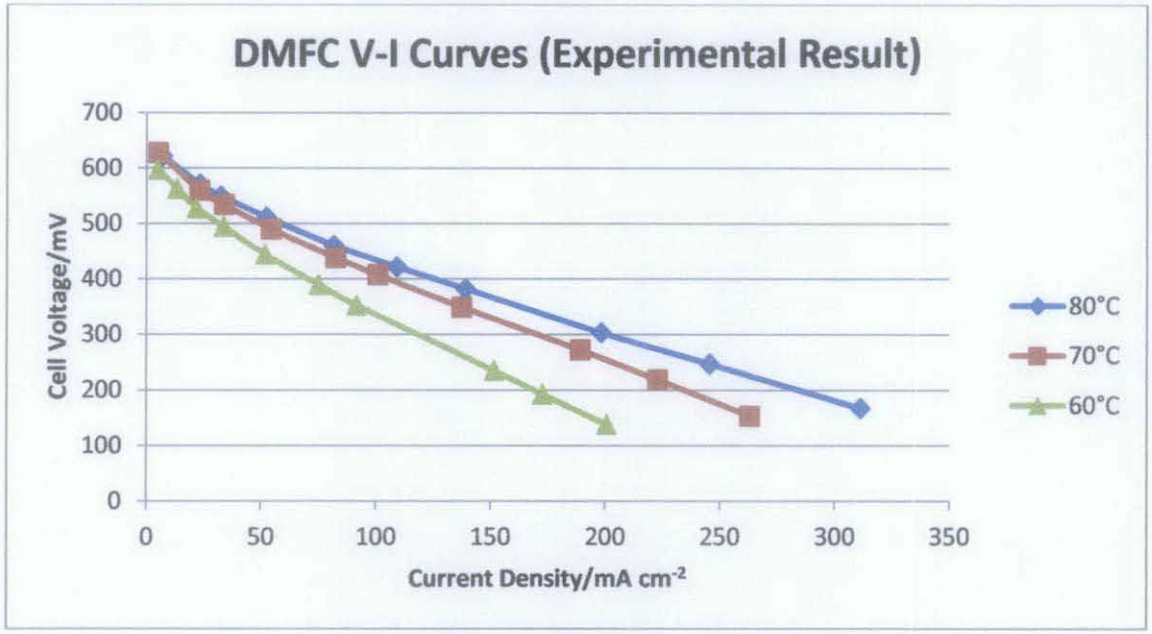


Figure 17: Effect of operating temperature on DMFC V-I curve (experimental result)

The result for simulation and experimental data is compared at 70°C. The graph is plotted as below:

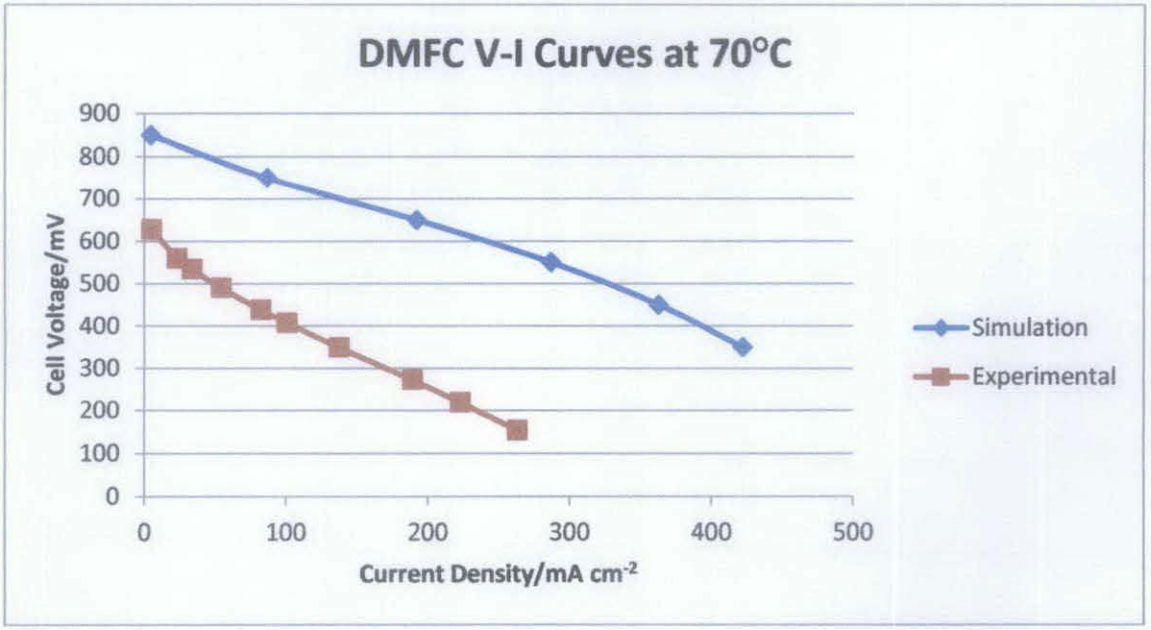
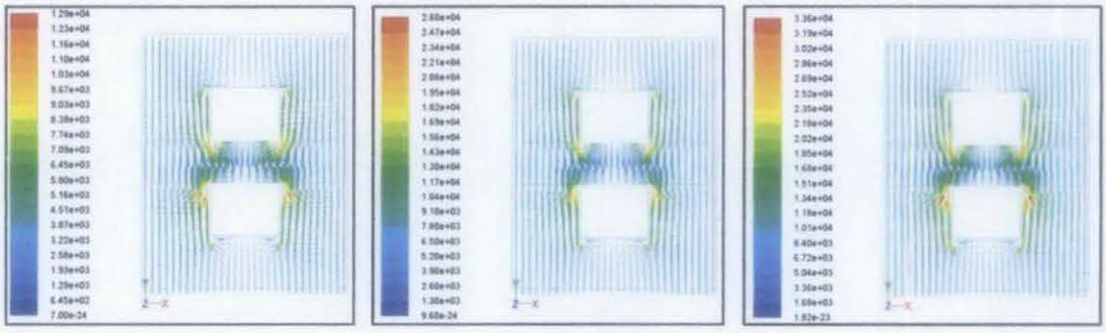


Figure 18: DMFC V-I curve at 70°C (simulation vs. experimental result)

Based on the results (simulation and experiment), both of them show a decreasing trend. It is notice that for a particular temperature (e.g. 70°C), the experimental data has a lower value that the simulation result. This kind of difference is expected as in the real experiment, the result is influenced by many other factors such as heat management, sluggish catalyst of methanol oxidation as well as the methanol crossover. While in the simulation, it only computed the effect of the methanol crossover on the fuel cell.

4.5 Data Analysis

For the first part, the result of the simulation is represented in the form of contour. Red colour shows the maximum value while blue colour is the minimum. The value that is shown in the contour represents the flux current-density. Comparing the 3 simulations with a constant cell voltage (750mV), the result shows that the maximum current density that the fuel cell needed is increasing with the temperature value.



60°C 70°C 80°C

Figure 19: Current flux density distribution, amp/m^2

The second part of the analysis is to construct a V-I curve to show the relationship between current density and cell voltage. The graph is plotted with the 3 different operating temperatures which are 60°C, 70°C and 80°C.

Overall, the graph matches the trends in the experimental result where it shows a decreasing cell voltage with an increasing current density value. Methanol crossover significantly increases the cathode over potential at low current density but its effect is reduced when the current density is increased to a certain amount of value.

From figure 17, as the temperature increased the value of current density is also increased in order to obtained a specific amount of cell voltage.

Temperature	Current Density at 450mV
60°C	303
70°C	363
80°C	398

This behaviour indicates that methanol crossover has influenced the efficiency of the fuel cell. The increased methanol crossover rate due to the temperature increment could be attributed to the easily transportation of methanol molecules through the polymer membrane resulting from their faster mobility because of the higher temperature. Another probably reason for this is due to the fact that at higher temperature the polymer backbone expands due to softening of the fluorinated chain

The electrochemical consumption of methanol in the anodic catalyst layer is a function of current density. Due to the formation of mixed potentials, caused by methanol permeation, the cathode potential is influenced to such an extent that the increase of the potential on the anode side of the cell can be overcompensated by the reduction of the potential in the case of the cathode overpotential.

Overall, a result has been obtained at this point of study with only observing at the temperature perspective and neglecting other factors that influenced the fuel cell efficiency. It can be concluded that the optimum temperature between these three values is the lowest one which is 60°C.

CHAPTER 5
CONCLUSION AND RECOMMENDATION

5.1 Conclusion

This project is about computational fluid dynamics modeling and simulation of methanol cross-over in direct methanol fuel cell. In FYP 1, all the preliminary studies regarding the topic have been done. The objectives, problem statement and methodology have been justified and it is relevant to the study. Journals and experimental data have been collected for the validation of the simulation later.

In FYP 2, this project has been started by doing some research study on the software that will be used. After that, the modeling and simulation of the methanol cross-over effect based on the selected journal will be done. The geometry and mesh has been successfully created for the model. The main part of the project will be the numerical model setup and simulation of the fuel cell.

To conclude, this project has successfully developed the model and simulating a single channel direct methanol fuel cell (DMFC) by using Computational Fluid Dynamics. It is understand that the operating temperature has given a major effect in the permeation of methanol through fuel cell membrane. Thus, it affects the overall efficiency of the fuel cell. The computational results indicate that the performance of fuel cell is relatively decreased with an increasing of operating temperature.

5.2 Recommendation

Many improvements can be done for future works and analysis. One of them is to make a CFD simulation and study on a different type of fuel cell geometry. The variable parameters that affect the methanol crossover can also be increased such as the effect of fuel concentrations, inlet fuel velocity, operating pressure and much more. Lastly, other software aside FLUENT should be given a try to model DMFC. It is appreciated if more researches can do a study on this Direct Methanol Fuel Cell as it has a very big potential to stand tall in the market as a competitive environmentally friendly power source generator.

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FYP 1 GANTT CHART

No	Detail / Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14
1	Selection of Project Topic	■	■												
2	Preliminary Research Work		■	■	■	■									
3	FYP1 Briefing		■	■											
4	Submission of Extended Proposal						■								
5	Proposal Defence								■	■					
6	Project Work							■	■	■	■	■	■		
7	Submission of Interim Draft Report													■	
8	Submission of Interim Report														■

FYP 2 GANTT CHART

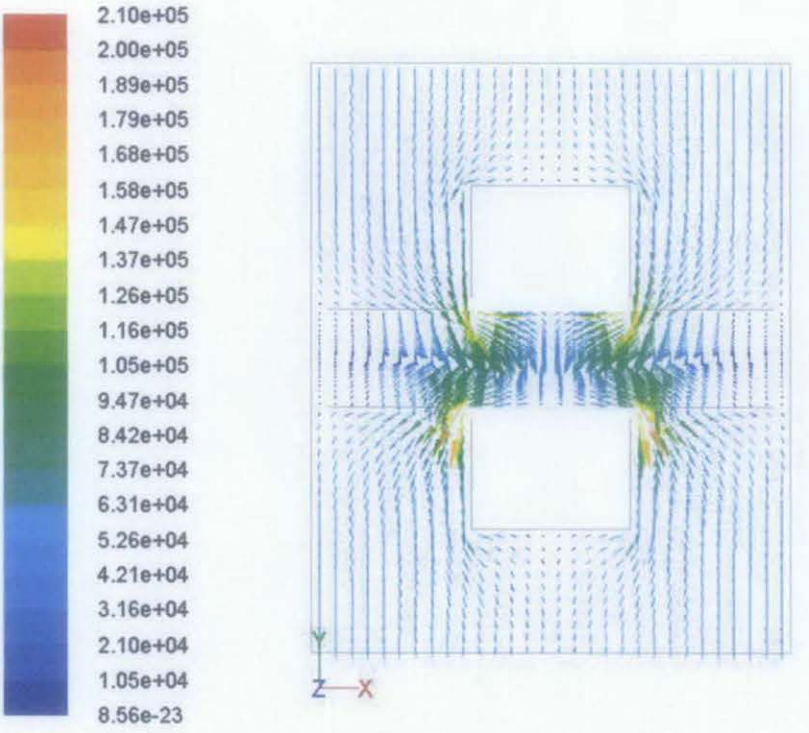
No	Detail / Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
1	Project Work Continues (FLUENT modeling)															
2	Submission of Progress Report															
3	Project Work Continues (FLUENT modeling and data validation)															
4	Pre-EDX															
5	Submission of Draft Report															
6	Submission of Dissertation (soft bound)															
7	Submission of Technical Paper															
8	Oral Presentation															
9	Submission of Project Dissertation (hard bound)															

APPENDIX B

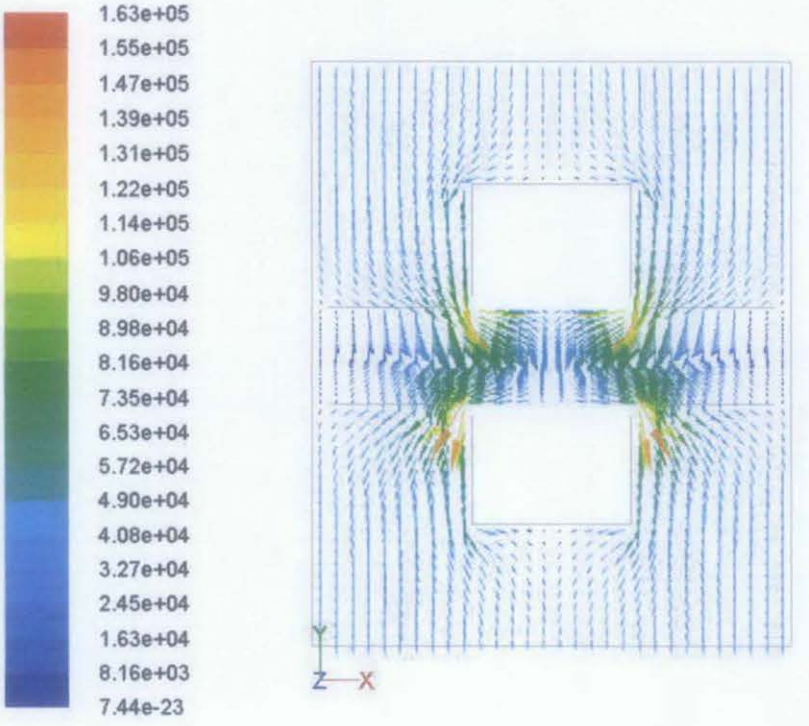
SIMULATION RESULTS

(CURRENT FLUX DENSITY DISTRIBUTION)

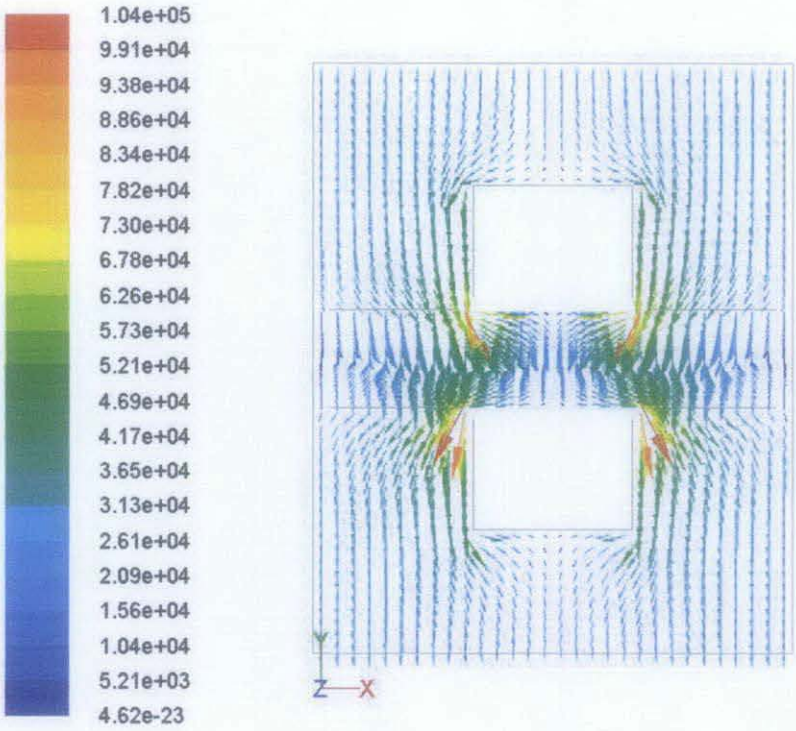
T = 60°C



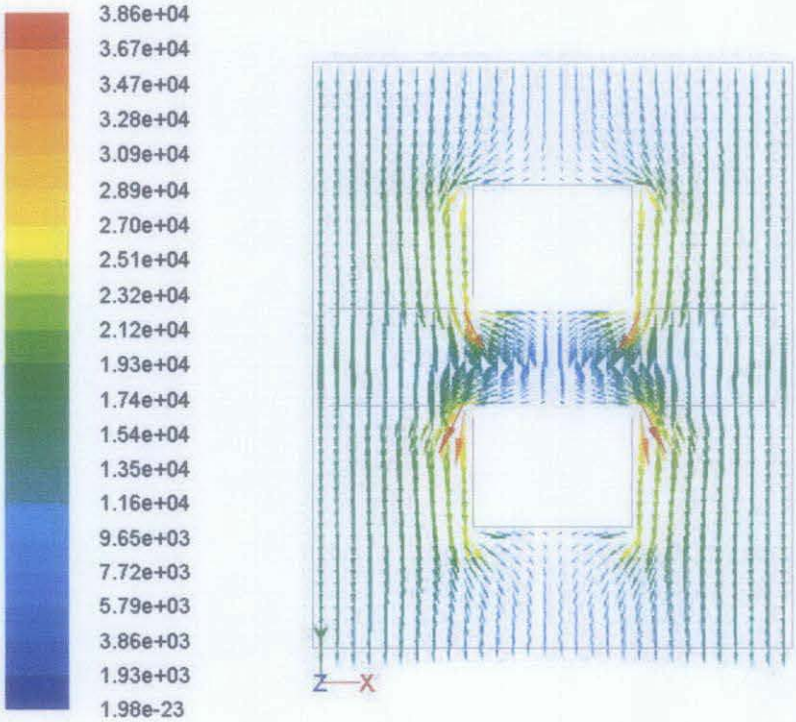
Cell Voltage = 350mV



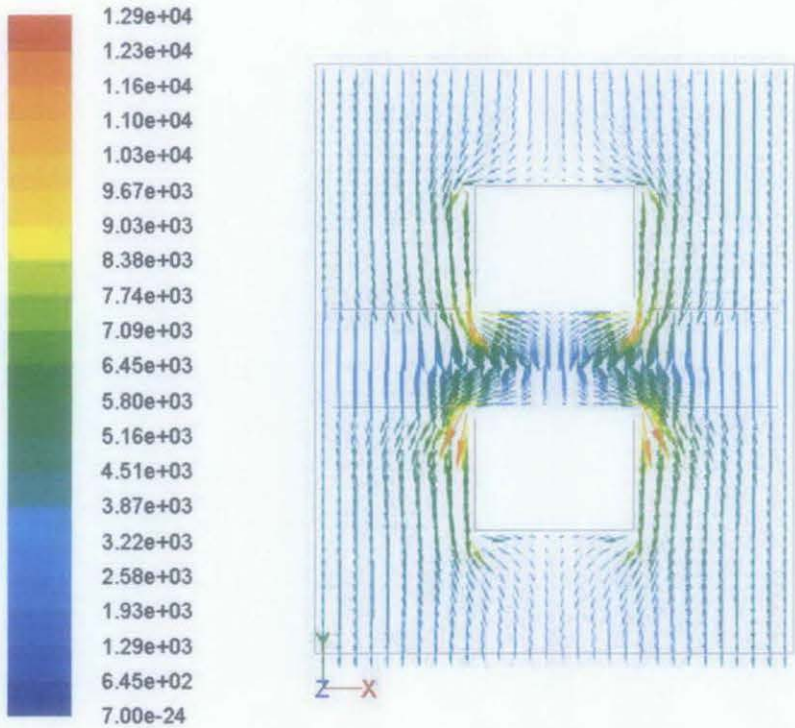
Cell Voltage = 450mV



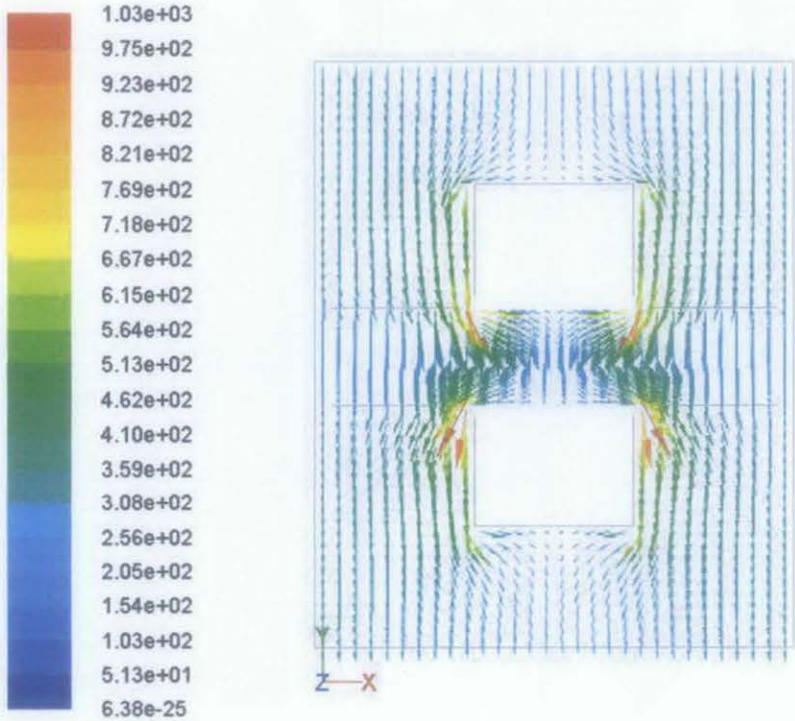
Cell Voltage = 550mV



Cell Voltage = 650mV

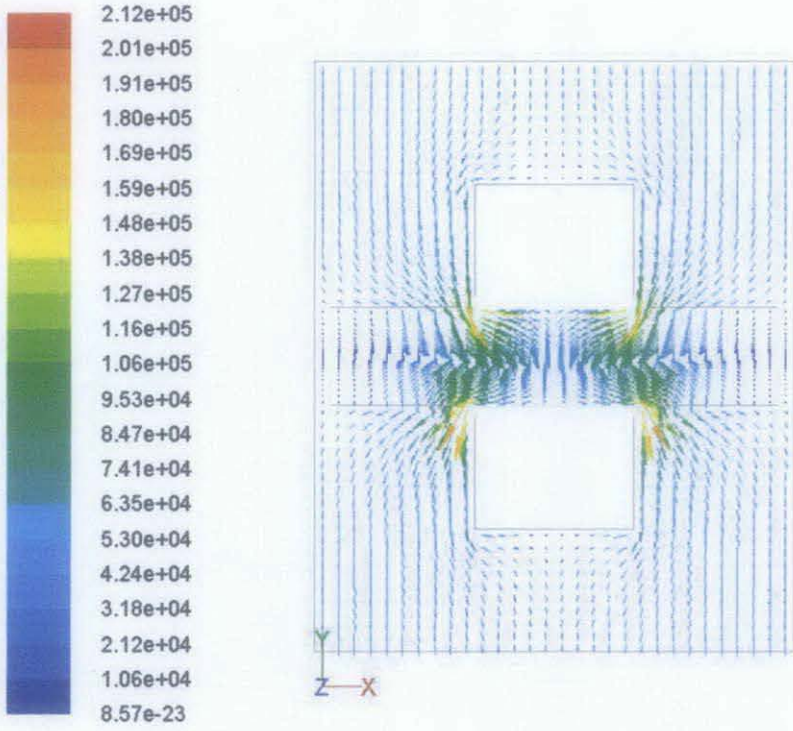


Cell Voltage = 750mV

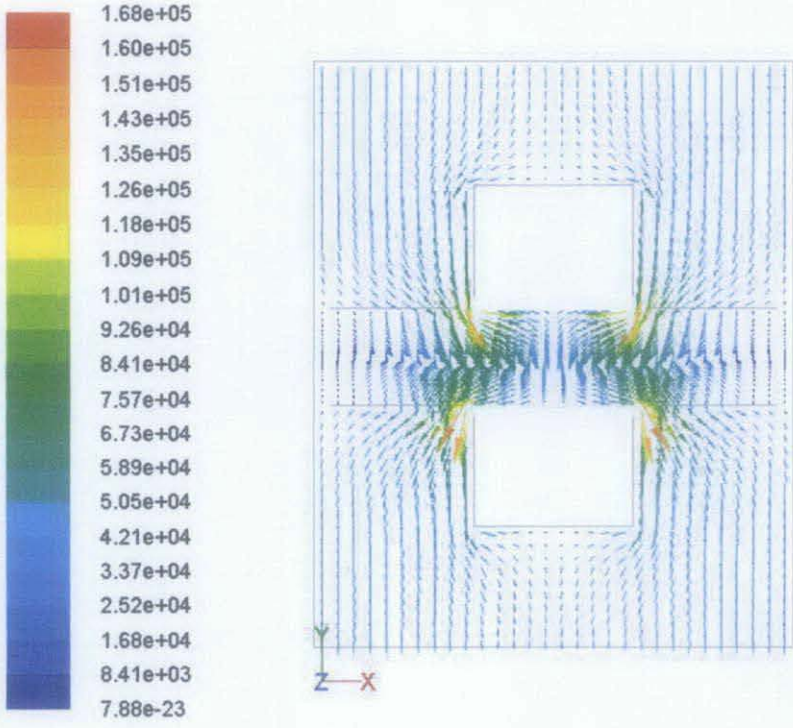


Cell Voltage = 850mV

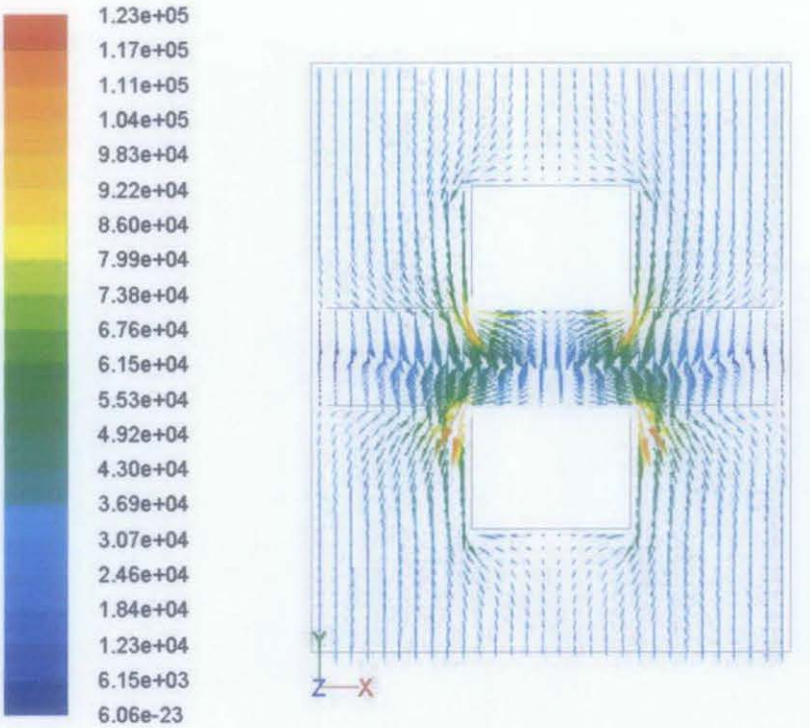
T = 70°C



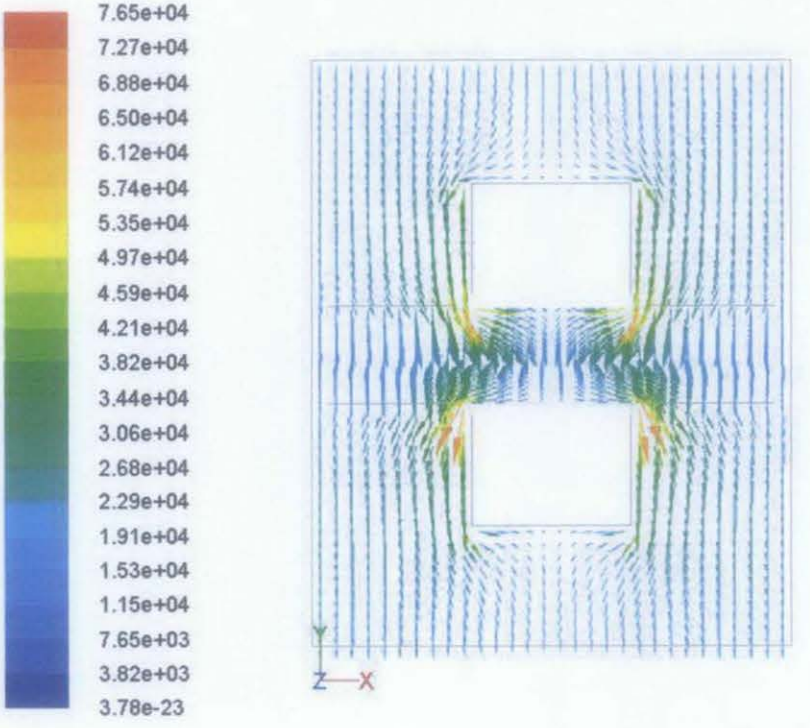
Cell Voltage = 350mV



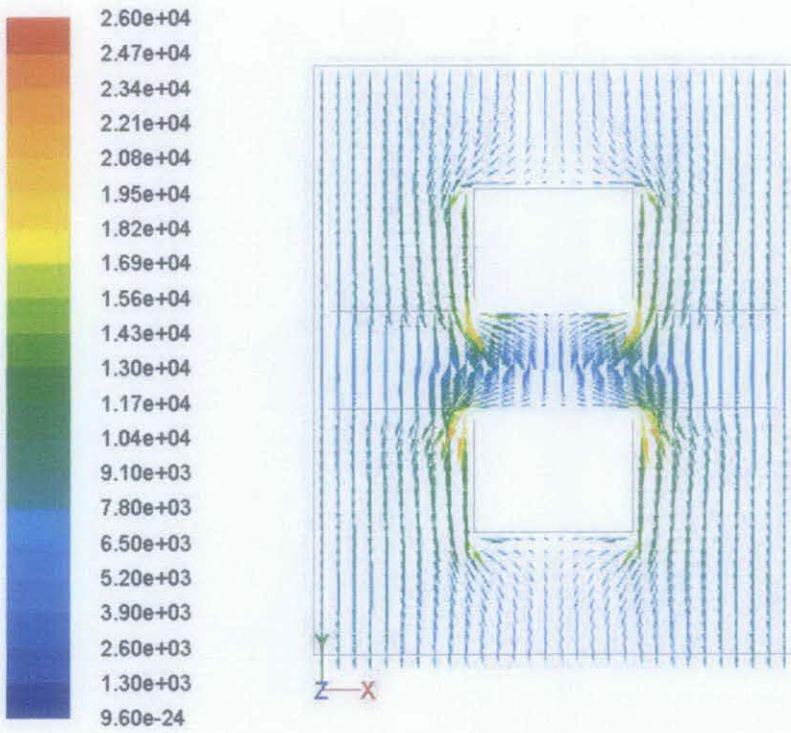
Cell Voltage = 450mV



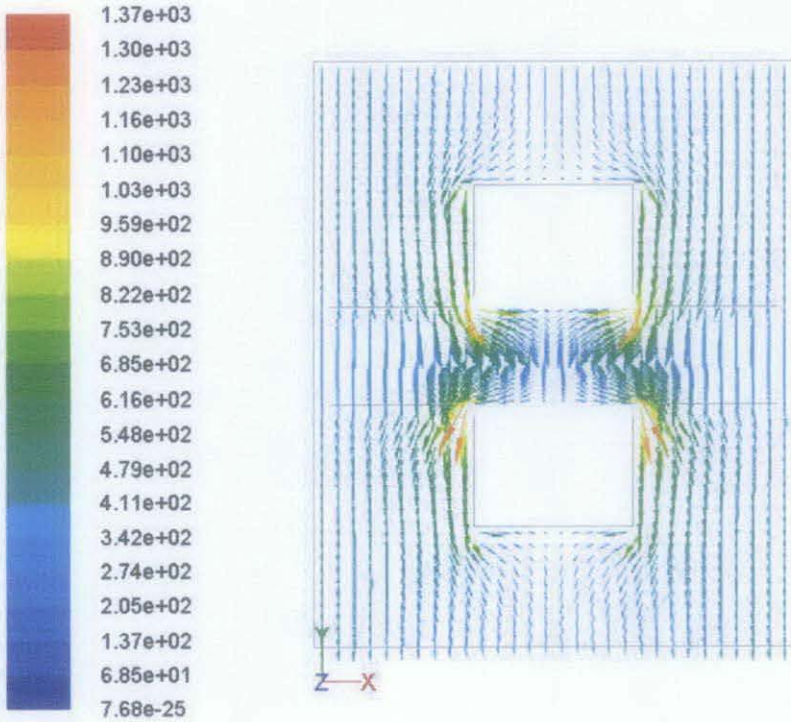
Cell Voltage = 550mV



Cell Voltage = 650mV

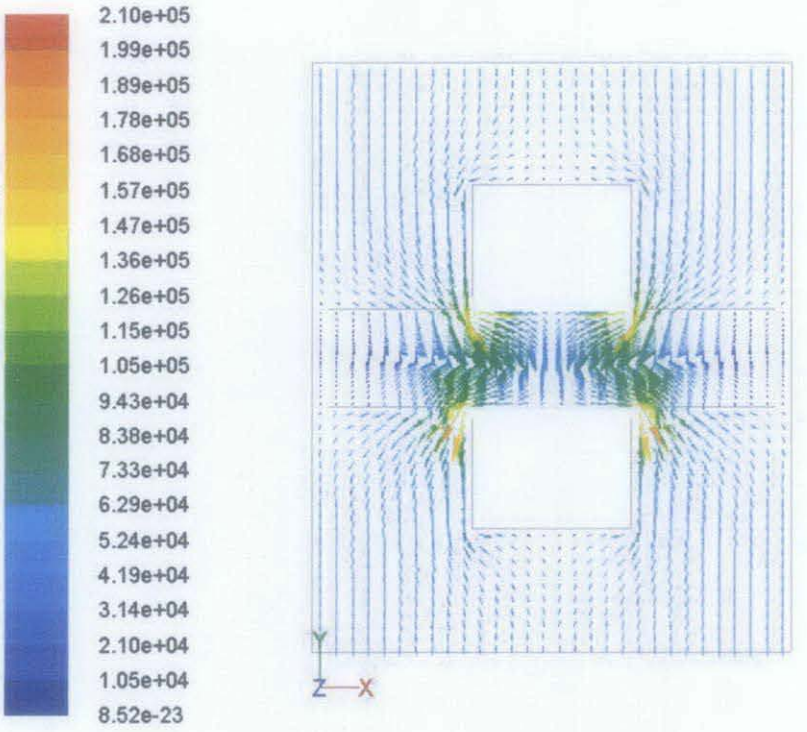


Cell Voltage = 750mV

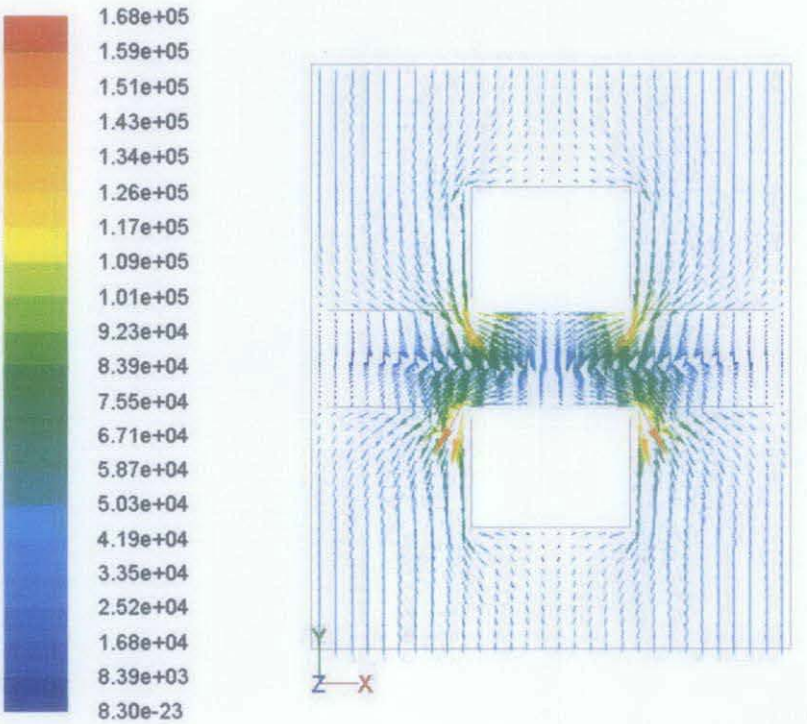


Cell Voltage = 850mV

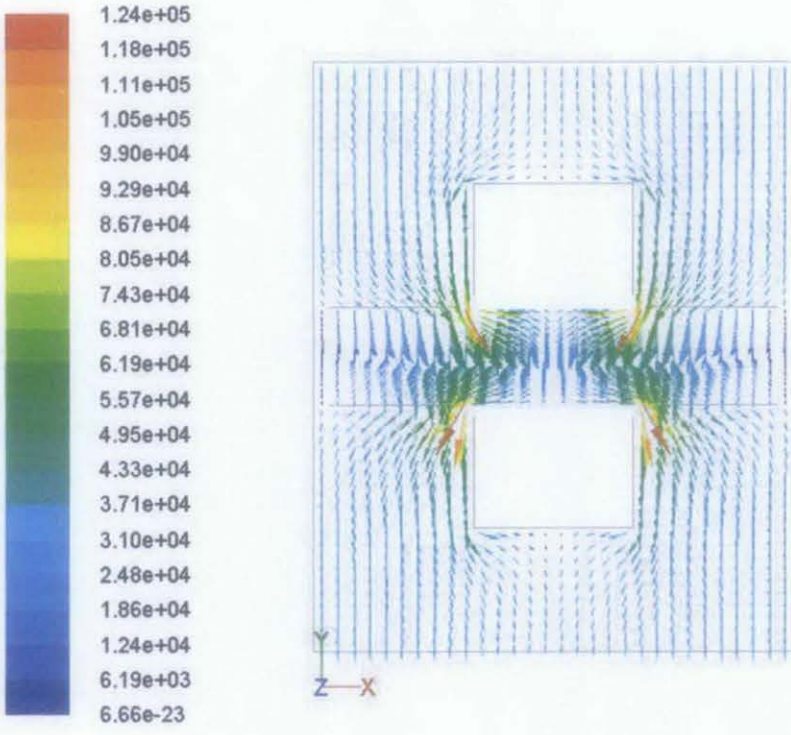
T = 80°C



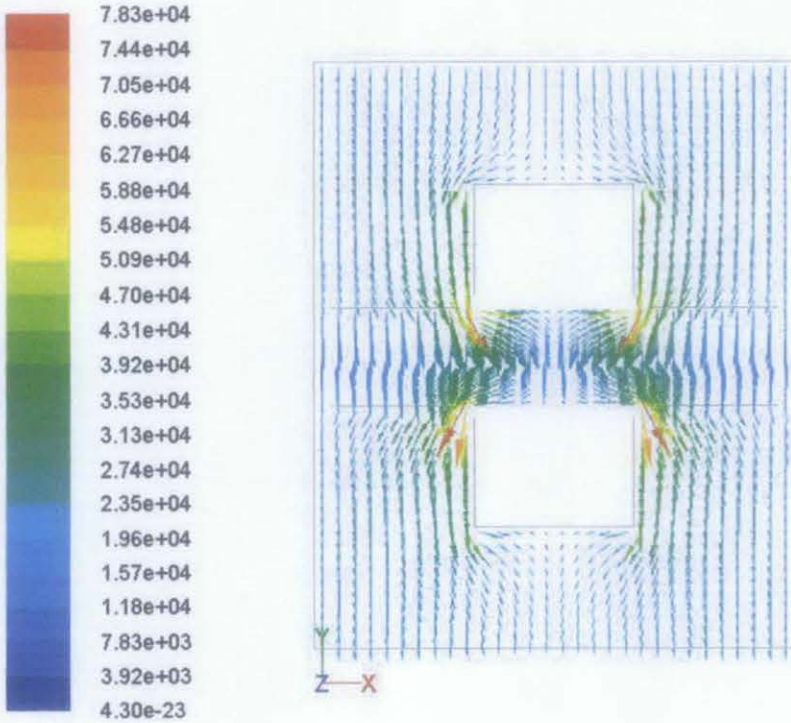
Cell Voltage = 350mV



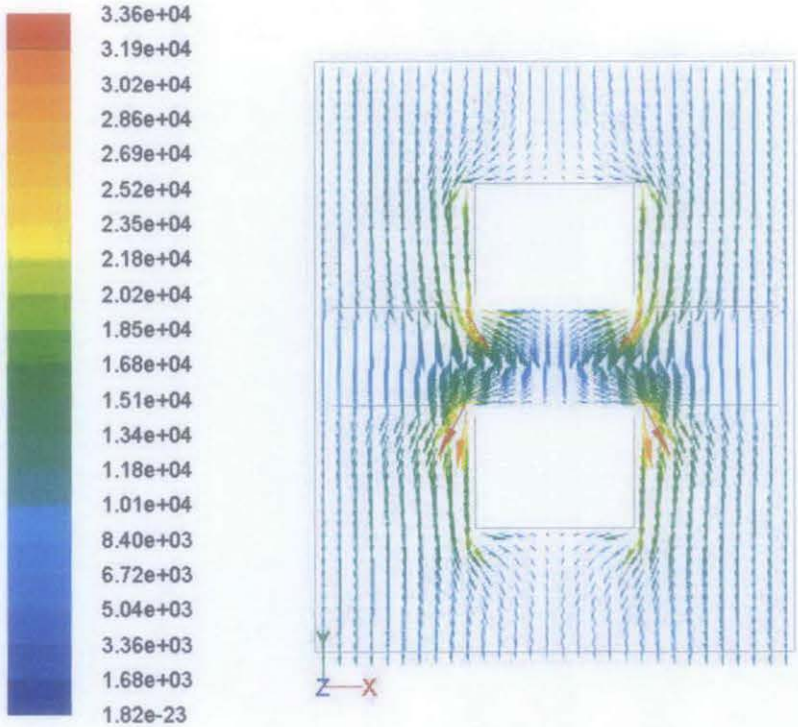
Cell Voltage = 450mV



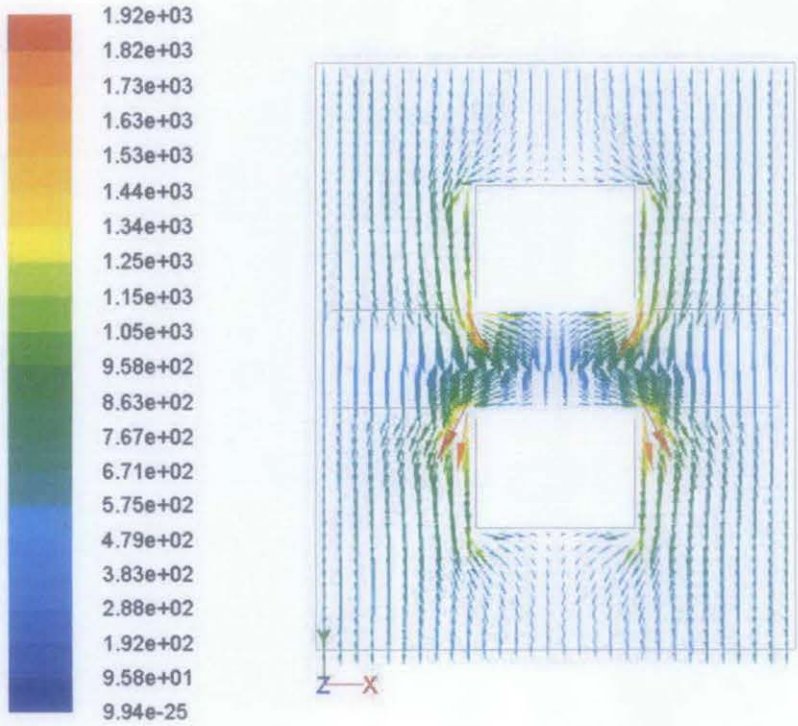
Cell Voltage = 550mV



Cell Voltage = 650mV



Cell Voltage = 750mV



Cell Voltage = 850mV

APPENDIX C

RESULT DATA FOR FLUENT SIMULATION

T = 60°C

Current Density (mA/cm^2)	Cell Voltage (mV)
4	850
61	750
146	650
229	550
303	450
349	350

T = 70°C

Current Density (mA/cm^2)	Cell Voltage (mV)
5	850
87	750
192	650
287	550
363	450
422	350

T = 80°C

Current Density (mA/cm^2)	Cell Voltage (mV)
7	850
118	750
230	650
322	550
398	450
461	350