A COMPUTATIONAL FLUID DYNAMIC FRAMEWORK FOR MODELING AND SIMULATION OF PROTON EXCHANGE MEMBRANE FUEL CELL

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A COMPUTATIONAL FLUID DYNAMIC FRAMEWORK FOR MODELING AND SIMULATION OF PROTON EXCHANGE MEMBRANE FUEL CELL

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy (Chemical Engineering)

Faculty of Chemical Engineering
Universiti Teknologi Malaysia

MAY 2015

To my beloved parents

ACKNOWLEDGEMENT

I would like to gratefully and sincerely appreciate my supervisor Dr. Mohd. Kamaruddin Abd. Hamid on her support, understanding, and motivation during my PhD study. Her guidance helped me in all the time of the research and writing of this thesis.

ABSTRACT

This thesis describes the development and application of a framework for model and analysis of proton exchange membrane fuel cells (PEMFCs) using computational fluid dynamics (CFD). The developed framework addresses the formulation, solution, and analysis of the PEMFCs systems in a systematic manner. This PEMFCs modelling framework helps to generate problem-system specific models describing a step by step proton exchange membrane (PEM) fuel cell model. Accordingly, the problem-system specific model generation procedure consists of three main steps. In the first step the problems and scope of the study are defined. The PEM fuel cell modelling procedure is done in the next step 2. This second step contains three sub-sections which are geometry definition, model definition and numerical solution and validation. In the step 3, the developed model is validated using available experiment/industrial data. A general, three-dimensional, non-isothermal, multi-phase numerical model has been developed to simulate and examine the fluid flow, heat and mass transfer, species transport, electrochemical reaction, and current density distribution of a PEMFC. The validation results of the PEM fuel cell model developed by using this framework has been successfully done. In addition, applications of the validated model with respect to advanced grid analysis, anisotropic properties investigation, and PEMFCs electrochemistry parameter have been successfully implemented. With respect to grid analysis, the results have shown that grid independence analysis using polarization curve is not accurate, where the concentration of fuel cell reactants and product showed more sensitivity for checking the grid independency. In terms of anisotropic properties investigation, the results have shown that increasing the value of anisotropy in thermal conductivity mitigates the gradient of liquid water between the area underneath the ribs and channels in PEMFCs. With respect to PEMFCs electrochemistry parameter, it has been shown that the new derived of the Kazemi-Jahandideh (K-J) approximation is able to reduce the numerical calculation to find electrochemistry parameters with a higher accuracy.

ABSTRAK

Tesis ini menerangkan pembangunan dan aplikasi rangka kerja untuk model dan analisa sel bahan api membran pertukaran proton (PEMFCs) menggunakan dinamik bendalir berkomputer (CFD). Rangka kerja ini telah dibangunkan untuk menangani masalah formula, penyelesaian, dan analisa sistem PEMFCs secara sistematik. Rangka kerja model PEMFCs dapat menghasilkan model khusus terhadap masalah-masalah sistem yang wujud dengan menerangkan secara terperinci bagi model sel bahan api membran pertukaran proton (PEM). Masalah-sistem terhadap prosedur penghasilan model adalah terdiri daripada tiga langkah utama. Dalam langkah pertama, masalah dan skop kajian ini telah ditakrifkan. Prosedur model bagi sel bahan api PEM telah dijalankan pada langkah kedua. Prosedur ini mengandungi tiga sub-bahagian iaitu definisi geometri, definisi model dan penyelesaian berangka dan pengesahan. Dalam langkah ketiga, model yang dibangunkan telah disahkan berdasarkan data eksperimen/ industri. Model simulasi yang umum, tiga dimensi, bukan isoterma dan mempunyai pelbagai fasa model berangka telah dibina untuk memeriksa aliran cecair, pemindahan haba dan jisim, pengangkutan spesies, tindakbalas elektrokimia, dan kepadatan taburan bagi PEMFC. Pengesahan rangka kerja terhadap model sel bahan api PEM telah berjaya dibangunkan. Selain itu, model aplikasi analisis grid, penyelidikan sifat anisotropik, dan parameter elektrokimia PEMFCs telah berjaya dilaksanakan. Berkenaan dengan analisis grid, keputusan telah menunjukkan bahawa analisis kebebasan grid menggunakan keluk polarisasi adalah tidak tepat, di mana kepekatan bahan tindak balas sel bahan api dan produk menunjukkan kesan sensitiviti terhadap kebebasan grid. Dari segi penyeldikan sifat anistropik, keputusan kajian telah menunjukkan bahawa peningkatan nilai anisotropik dalam konduksi terma dapat mengurangkan kecerunan cecair di antara kawasan tulang rusuk dan saluran dalam PEMFCs. Berkenaan PEMFCs parameter elektrokimia PEMFC, ia juga telah menunjukkan bahawa anggaran baru Kazemi-Jahandideh (K-J) dapat mengurangkan pengiraan berangka untuk mencari parameter elektrokimia dengan ketepatan yang lebih tinggi.

TABLE OF CONTENTS

LIST OF ABBREVIATIONS

CAD - Computer Aided Design
CFD - Computational Fluid Dynamic
CL - Catalyst Layer
EIS - Electrochemical Impedance Spectroscopy
GDL - Gas Diffusion Layer
HOR - Hydrogen Oxidation Reaction
ORR - Oxygen Reduction Reaction
PEM - Proton Exchange Membrane
PEMFC - Proton Exchange Membrane Fuel Cell

LIST OF SYMBOLS

a - Water Activity

A_{ch} - Gas Channel Cross-Sectional Area

A_m - Membrane Active Area

C_i - Concentration of Specie i

Cp - Heat Capacity

$-D$ - Dimensional

D^{eff} - Effective Diffusion Coefficient

e^- - Electron

E_0 - Cell Thermodynamic Potential

F - Faraday Constant

H^+ - Proton

i - Current Density

i_0 - Exchange Current Density

I_{cell} - Cell Current Density

k^{eff} - Effective Thermal Conductivity

M - Molar Mass

n - Electron Number

n_d - Electro-Osmotic Drag Coefficient

P - Pressure

R - Gas Constant

R_m - Membrane Resistance

S_e - Source Term/Sink of Electrical Conservation Equation

S_g - Source Term/Sink of Mass Conservation Equation

S_i - Source Term/Sink of Ionic Conservation Equation

S_l - Source Term/Sink of Liquid Water

S_m - Source Term/Sink of Momentum Conservation Equation

S_{phase} - Source Term/Sink of Water in Corresponding Phase

S_T - Source Term/Sink of Energy Conservation Equation

S_λ - Source Term/Sink of Dissolved Water

T - Temperature

V - Velocity
V_{cell} - Cell Potential
w - Mass Fraction

Greek letters

β - Symmetry Factor
α - Transfer Coefficient
ε - Porosity
σ Electrical Conductivity
η - Overpotential
μ - Viscosity
ζ Stoichiometric Ratio
Φ_i - Ionic Potential
Φ_e - Electrical Potential
λ - Membrane Water Content

Subscripts

act - Activation
a - Anode
c - Cathode
$conc.$ - Concentration
$resist$ - Resistance
g - Gas Phase
i - Specie
$cell$ - Fuel Cell

Superscripts

ref - Reference
eff - Effective

LIST OF TABLES

LIST OF FIGURES

CHAPTER 1

INTRODUCTION

1.1 Introduction

Fuel cells are future technology as a major energy conversion and there are many electrochemical advantages conversion processes in compared with thermal combustion processes (Scherer and Gürsel, 2008). Fuel cells share the electrochemical nature of the power generation process same to batteries but unlike batteries fuel cells work continuously consuming a fuel of different type. A fuel cell defined as an device that changes over compound vitality into electric vitality electrochemically continuously until fuel and oxidant are supplied (Hoogers, 2002).

Fuel cells are clean devices because of its by-products (for example, water when fuel cell fuel is pure hydrogen) and they also operate quietly because they do not have any moving parts. Moreover, they have high power density and effectiveness, commonly more than 40% efficiency in electric power production, which is better than sets of traditional combustion (Gou *et al.*, 2009).

The most common fuel for fuel cells is hydrogen, which may be supplied as almost pure hydrogen or as a major component (50 to 70%) of a reformed fuel stream. Alternatively, a hydrogen-containing component may be used, such as methanol in a direct methanol fuel cell (Li, H. *et al.*, 2010).

Three distinct fuel cell technologies currently exist. Two types are well proven for providing electrolytic alkaline and PEM fuel cell. In general, the PEM fuel cell offers several advantages compared with alkaline fuel cell much higher current densities; smaller mass and volume characteristics; high degree of purity; increased level of safety (no circulation of caustic electrolyte); possibility of combining a fuel cell with electrolyzer modes. A third type usually termed as solid-oxide fuel cell (SOFC), is currently under development and may be viable in the longer term. Practical SOFC operates at high temperature (800 C). They are properly designed to operate in combination with nuclear plants or other technologies where there is an extra amount of heat at a low cost. In safety-critical applications, PEM fuel cell reliability has been extremely high, with units achieving 100,000 h operation without failure (Siracusano *et al.*, 2012).

PEM fuel cell/electrolyzer used in a photovoltaic array to save green energy (Figure 1.1). A photovoltaic array drives a PEM (Proton-Exchange Membrane) fuel cell, producing on-demand electricity to power a load. The only required input is energy to drive the electrolyzer-the water and gases cycle in a closed loop (Shapiro *et al.*, 2005).

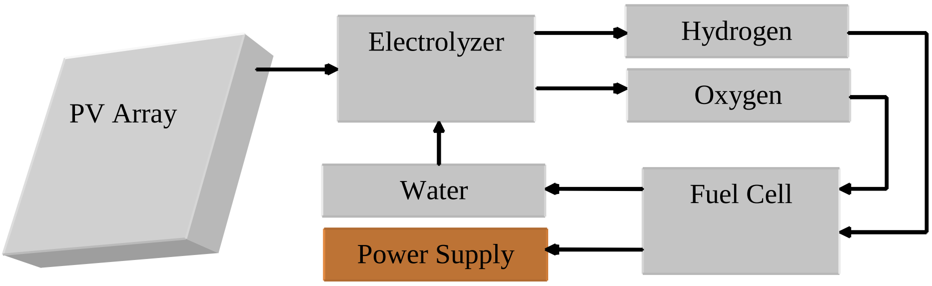

Figure 1.1 Photovoltaic electrolyzer/fuel cell system

The PEM fuel cell schematic is presented in figure 1.2. The PEM fuel cell consists of the solid membrane with gas diffusion layers and the anode and cathode electrodes. An anode and cathode electrode consists of catalyst layer particles, and is fixed to the gas diffusion layer or solid membrane. porous and electrically conductive substance have been used to made the gas diffusion layer , such as carbon cloth, to permeate the fuel feed to diffuse into and out of the PEM, and to collect the resulting current by providing electric contact between the electrode and the outside bipolar plate. Moreover, it allows the formed water at the cathode electrode to exit to the cathode gas channels. The bipolar plates, also called flow field plates, distribute the reactant gas over the surface of the electrodes through flow channels on their surfaces. (Gou *et al.*, 2009). The electrochemical reactions that happen on both sides of membrane in fuel cell– the anode and cathode electrods are (Kakaç *et al.*, 2008):

At the anode side : $H_2 \rightarrow 2H^+ + 2e^-$

At the cathode side : ${}^1\!/_2 O_2 + 2H^+ + 2e^- \rightarrow H_2O$

Overall: $H_2 + {}^1\!/_2 O_2 \rightarrow H_2O$

In a PEM fuel cell, both the anodic hydrogen (or liquid fuel) oxidation reaction (HOR) and the cathodic oxygen reduction reaction (ORR) take place within the respective catalyst layers. Electrocatalysts and their corresponding catalyst layers, thus play critical roles in fuel cell performance. In our present state of technology, the most practical catalysts in PEM fuel cells are highly dispersed Pt-based nanoparticles (Zhang, Jiujun, 2008). The heart of the PEM fuel cell is the proton exchange membrane, which transports protons from the anode to the cathode. The membrane also serves to separate the fuel and oxidant gas phases and electronically insulates the cathode from the anode. The most typical membrane is a sulfonated perfluorinated polymer. The Nafion® family of membranes made by DuPont is representative of this class, and is based on a sulfonated tetrafluoroethylene based fluoropolymer-copolymer (Li, H. *et al.*, 2010).

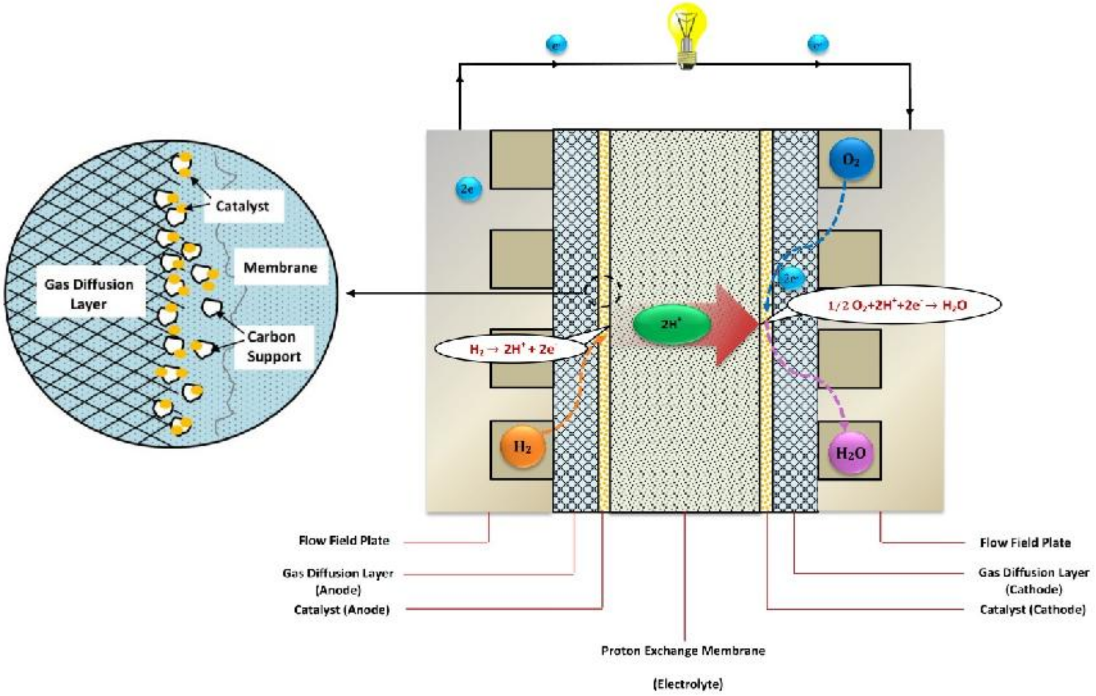

Figure 1.2 PEM fuel cell schematic

1.2 Problem Statements

Increasing the efficiency is the basic reason that modeling and design optimization of PEM fuel cell need to be performed. Several researches by different method and tools have been done to improve the PEM fuel cell but there are still some gap that needed more study. Although many studies available in the literature but simple types of modeling did not have the ability to consider all aspect of cells and stacks development in one single framework (Selamet *et al.*, 2011). The comprehensive model is also very complex. CFD is an emerging technique to investigate the PEMFCs systems. In this situation CFD modeling is close to the real condition and make helpful and easy for scaling-up. According to this literature review, following aspects of CFD modeling of PEM fuel cell problem need to be considered and solved:

Most of modeling domain, such as analytical and semi-empirical do not have the ability to analyze the geometry of the cell. On the other hand, there is no updated framework for mechanistic modeling.

i. CFD modeling has a potential to be used to investigate the cell geometry since CFD is a powerful tool to model the geometry analysis with high accuracy but there is an doubt on geometry analysis in CFD modelling.
ii. Flooding is one of the most important phenomena that has a bad effect on PEM fuel cell performance and durability. However, analytical modelling does not have the ability to investigate this phenomenon. CFD modelling has a potential to evaluate the flooding phenomena on the anisotropic gas diffusion layer properties.
iii. One of the disadvantages of CFD is the calculation time because of solving a set of differential conservation equations in each of the computational zones. Finding the new axillary relations may improve this limitation. The modification of a new approximation to approximate some of the PEMFCs parameters such as electrochemistry properties has a potential in resolving the limitation of the CFD.

1.3 Objective of the Study

The main objective of this study is to develop a systematic computational fluid dynamic modeling framework of PEM fuel cell for electricity generation. These specific objectives were constructed in order to achieve the main objective:

i. To increase the geometry analysis accuracy of the PEM fuel cell system through grid independence test.
ii. To evaluate the anisotropic property effects over flooding phenomenon in the PEM fuel cell system.
iii. To improve the prediction of electrochemistry property relation in a fuel cell system.

1.4 Scope of the Work

In order to achieve the above mentioned objectives, the following scope of work was proposed:

The PEM fuel cell CFD modelling could be conducted in the channel, serpentine (a U type channel), cell or stack system, but in this research it was only conducted for the serpentine system of a cell. Investigations in the serpentine system where the fundamental investigation of PEM FC research. A great potential extended investigations for a cell or a stack system will be easier if these fundamental investigations can successfully be achieved.

i. An investigation of the geometry analysis accuracy was only applied to the grid independence test. Grid independence test is important because the fuel cell has a simple geometry and focusing on the other aspect of geometry such as mesh adapting will be not necessary.

ii. The anisotropic properties effect on the flooding phenomenon has been investigated only in a gas diffusion layer of the cathode. This investigation is important because, the flooding only happened on the cathode side of the cell and catalyst layer anisotropic properties effect are negligible due to this layer is very thin in thickness.
iii. The activation overpotential is the electrochemistry property that this research focused to improve the approximation to predict it. Simple approximation can reduce the numerical calculation time in CFD modelling of PEM fuel cell.

1.5 Organization of Thesis

The thesis consists of 7 chapters, chapter 1 outline a brief introduction of PEM fuel cell and background of the research. Then, it is followed by the problem statements, which identify the research direction. Based on the problem statements defined, the objectives and scopes of the elaborate study in details. General overview of different modeling of a PEM fuel cell is presented in chapter 2. The review of thesis objectives also has been presented in chapter two. Chapter 3 describes the computational fluid dynamics methodology for PEM fuel cell modeling. In this chapter the theory behind the electrochemistry modeling, current and mass conservation, liquid water formation and transport phenomena are presented. The results of PEM fuel cell validation have been achieved through the CFD modeling also is presented in chapter 3. Other results according to research objectives are presented in chapter 4 to 6, respectively. Finally, the general conclusion is drawn from this research and some recommendations for the future research are provided in chapter 7.

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