The influence of active area and stacking on PEM fuel cell performance: A simulation modelling and experimental investigation

A thesis submitted in fulfilment of the requirements for the degree of Doctor of Philosophy

Xiang Wu

School of Engineering
College of Science, Engineering and Health
RMIT University

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DECLARATION

I, Xiang Wu, hereby submit the thesis entitled “The influence of active area and stacking on PEM fuel cell performance: A simulation modelling and experimental investigation” for the degree of Doctor of Philosophy and certify that except where due acknowledgement has been made, the work is that of the author alone; the work has not been submitted previously, in whole or in part, for any other academic award and that the content of the thesis is the result of work that has been carried out since the official commencement of the program.

Xiang Wu

31 Mar 2017
Table of Contents

LIST OF ABBREVIATIONS ........................................................................................................... 7
NOMENCLATURE ........................................................................................................................... 9
LIST OF FIGURES .......................................................................................................................... 11
LIST OF TABLES ............................................................................................................................ 15
EXECUTIVE SUMMARY .............................................................................................................. 16
1. INTRODUCTION ....................................................................................................................... 21
   1.1 BACKGROUND .................................................................................................................... 21
   1.2 Research on Proton Exchange Membrane Fuel Cell ............................................................ 25
   1.3 AIM AND OBJECTIVES OF THIS THESIS ........................................................................ 27
   1.4 RESEARCH QUESTIONS ..................................................................................................... 27
   1.5 SCOPE .................................................................................................................................. 28
   1.6 DELIVERABLES .................................................................................................................... 29
   1.7 GUIDE TO THESIS .............................................................................................................. 29
2. METHOD .................................................................................................................................... 32
   2.1 OVERALL METHODOLOGICAL APPROACH ...................................................................... 32
      2.1.1 Computer simulation modelling ..................................................................................... 32
      2.1.2 Validating the ANSYS Fuel Cell Module against experiment ....................................... 33
      2.1.3 Applying ANSYS module to larger single fuel cell and fuel cell stack .......................... 35
      2.1.4 Limitations of this method ............................................................................................. 35
   2.2 MAIN PHASES AND ACTIVITIES ....................................................................................... 36
      2.2.1 Literature review ............................................................................................................ 36
      2.2.2 Application of the ANSYS Fuel Cell Module to a basic fuel cell with a single straight
            channel for each gas ........................................................................................................... 36
      2.2.3 Comparison of ANSYS Fuel Cell simulation modelling of small single fuel cells with
            experimental data ............................................................................................................. 37
      2.2.4 Investigation of the effects of membrane active area on a single large fuel cell
            performance ....................................................................................................................... 37
      2.2.5 Investigation of the Mass-transfer constrain in ANSYS Fuel Cell Module .................... 37
      2.2.6 Investigation of the difference between performance on fuel cell stacks and
            performance on single fuel cells ....................................................................................... 38
      2.2.7 Drawing conclusion and making recommendations ........................................................ 38
3. REVIEW OF PEM FUEL CELL HISTORY, SIMULATION MODELLING AND ITS
   EXPERIMENTAL VALIDATION ................................................................................................. 39
   3.1 PROTON EXCHANGE MEMBRANE FUEL CELLS .............................................................. 39
      3.1.1 Brief history of hydrogen fuel cells .................................................................................. 39
5.3.3 Cell C .................................................................................................................. 134
5.4 Conclusions .......................................................................................................... 141
6. SIMULATION OF A LARGE PEM FUEL CELL .................................................. 143
  6.1 INTRODUCTION ................................................................................................. 143
  6.2 ANSYS FUEL CELL MODULE SIMULATION OF THE SINGLE LARGE CELL ..... 143
    6.2.1 Geometry of single large fuel cell ............................................................... 143
    6.2.2 Mesh for simulation ..................................................................................... 145
    6.2.3 Simplification of geometry ......................................................................... 147
    6.2.4 Input settings ............................................................................................... 147
    6.2.5 Results and analysis ................................................................................... 148
    6.2.6 Comparison with experimental data ........................................................... 153
  6.3 Modelling the mass transfer constraint in a PEM fuel cell ............................. 155
    6.3.1 ANSYS simulation of mass transfer constraint for large single PEM cell .... 155
    6.3.2 How the mass transfer constraint is represented in the ANSYS Fuel Cell Module .... 156
    6.3.3 Input parameters affecting mass transfer constraint ............................... 164
  6.4 Conclusion ......................................................................................................... 170
7. SIMULATION MODELLING OF FUEL CELL STACKS IN THE ANSYS FUEL CELL
   MODULE ............................................................................................................... 172
  7.1 MODELLING FUEL CELL STACKS .................................................................. 172
  7.2 MODEL OF PEM FUEL CELL STACK ............................................................... 175
    7.2.1 Model setup ............................................................................................... 175
    7.2.2 Outputs and Analysis ............................................................................... 178
  7.3 Conclusions ....................................................................................................... 184
8. CONCLUSION AND RECOMMENDATIONS ..................................................... 186
  8.1 INTRODUCTION ............................................................................................... 186
  8.2 RESPONSE TO THE RESEARCH QUESTIONS ............................................ 186
    8.2.1 To what level of accuracy can simulations of the performance of individual PEM
         fuel cells using the ANSYS Fuel Cell Module match experimental performance data? .. 186
    8.2.2 How does changing the membrane active area affect the performance of a single PEM
         fuel cell in terms of current density and power density? ............................... 188
    8.2.3 How does the performance of a PEM fuel cell stack change compared to single fuel
         cells in terms of current density and power density? .................................... 191
    8.2.4 What is the best approach to using the ANSYS Fuel Cell Module to represent the
         mass-transfer limited portion of the voltage current density curve of a PEM fuel cell at higher
         current densities? ......................................................................................... 194
  8.3 CONCLUSIONS ................................................................................................. 195
  8.4 RECOMMENDATIONS ...................................................................................... 196
REFERENCES .................................................................................................................. 198
Publications arising from this thesis .................................................................................. 204
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>3D</td>
<td>Three dimensional</td>
</tr>
<tr>
<td>A</td>
<td>Ampere</td>
</tr>
<tr>
<td>a</td>
<td>Anode</td>
</tr>
<tr>
<td>AFC</td>
<td>Alkaline fuel cell</td>
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<tr>
<td>c</td>
<td>Cathode</td>
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<tr>
<td>CFD</td>
<td>Computational fluid dynamics</td>
</tr>
<tr>
<td>CHP</td>
<td>Combined heat and power</td>
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<tr>
<td>CL</td>
<td>Catalyst layer</td>
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<tr>
<td>cm</td>
<td>Centimeter</td>
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<tr>
<td>CO2</td>
<td>Carbon dioxide</td>
</tr>
<tr>
<td>CTD</td>
<td>Capability and technology demonstrator</td>
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<tr>
<td>DDA</td>
<td>Deacetylation</td>
</tr>
<tr>
<td>DMFC</td>
<td>Direct methanol fuel cell</td>
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<tr>
<td>E</td>
<td>Electrical</td>
</tr>
<tr>
<td>FCEV</td>
<td>Fuel cell electric vehicle</td>
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<tr>
<td>GDL</td>
<td>Gas diffusion layer</td>
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<tr>
<td>GE</td>
<td>General electric</td>
</tr>
<tr>
<td>HP</td>
<td>High performance</td>
</tr>
<tr>
<td>IFC</td>
<td>International fuel cells</td>
</tr>
<tr>
<td>K</td>
<td>Kelvin</td>
</tr>
<tr>
<td>kg</td>
<td>Kilogram</td>
</tr>
<tr>
<td>LB</td>
<td>Lattice boltzmann</td>
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<tr>
<td>LS</td>
<td>Level set</td>
</tr>
<tr>
<td>M²</td>
<td>Multi-phase mixture</td>
</tr>
<tr>
<td>MCFC</td>
<td>Molten carbonate fuel cell</td>
</tr>
<tr>
<td>MEA</td>
<td>Membrane electrode assembly</td>
</tr>
<tr>
<td>mm</td>
<td>Millimeter</td>
</tr>
<tr>
<td>Mt</td>
<td>Mega ton</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Full Form</td>
</tr>
<tr>
<td>--------------</td>
<td>-----------------------------------------------</td>
</tr>
<tr>
<td>Mtoe</td>
<td>Mega tone of oil equivalent</td>
</tr>
<tr>
<td>NASA</td>
<td>National Aeronautics and Space Administration</td>
</tr>
<tr>
<td>O₂</td>
<td>Oxygen</td>
</tr>
<tr>
<td>OPEC</td>
<td>Organization of the Petroleum Exporting Countries</td>
</tr>
<tr>
<td>ORR</td>
<td>Oxygen reduction reaction</td>
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<tr>
<td>PAFC</td>
<td>Phosphoric acid fuel cell</td>
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<tr>
<td>PEM</td>
<td>Proton exchange membrane</td>
</tr>
<tr>
<td>PN</td>
<td>Pore-network</td>
</tr>
<tr>
<td>Pt</td>
<td>Platinum</td>
</tr>
<tr>
<td>PTFE</td>
<td>Polytetrafluoroethylene</td>
</tr>
<tr>
<td>Ref</td>
<td>Reference</td>
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<tr>
<td>S</td>
<td>Sigma</td>
</tr>
<tr>
<td>SOFC</td>
<td>Solid oxide fuel cell</td>
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<tr>
<td>TCR</td>
<td>Thermal contact resistance</td>
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<tr>
<td>TPB</td>
<td>Triple phase boundary</td>
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<tr>
<td>TWh</td>
<td>Terawatt-hours</td>
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<tr>
<td>UDF</td>
<td>User defined functions</td>
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<tr>
<td>UDS</td>
<td>User defined scalar</td>
</tr>
<tr>
<td>URFC</td>
<td>Unitised regenerative fuel cell</td>
</tr>
<tr>
<td>VOF</td>
<td>Volume of fluid</td>
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<tr>
<td>W</td>
<td>Watt</td>
</tr>
<tr>
<td>y</td>
<td>Year</td>
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NOMENCLATURE

V \quad \text{Voltage} \\
J \quad \text{Current density} \\
\sigma \quad \text{Electrical conductivity} \\
\phi \quad \text{Electric potential} \\
R \quad \text{Volumetric transfer current} \\
\gamma \quad \text{Concentration dependence exponent} \\
\alpha \quad \text{Exchange coefficient} \\
F \quad \text{Faraday constant} \\
R \quad \text{Gas constant} \\
h \quad \text{Enthalpy change} \\
\gamma_{\text{an/cat}} \quad \text{Reference exchange current density per active surface area} \\
\zeta \quad \text{Specific active surface area} \\
[A] \quad \text{Local species concentration in anode side} \\
[A]_{\text{ref}} \quad \text{Reference species concentration in anode side} \\
\gamma \quad \text{Concentration dependence exponent} \\
\alpha \quad \text{Exchange coefficient} \\
s \quad \text{Water saturation} \\
r_{\text{w}} \quad \text{Condensation rate} \\
c_{\text{r}} \quad \text{Condensation rate constant} \\
V_{\text{l}} \quad \text{Liquid velocity} \\
\varepsilon \quad \text{Porosity} \\
\delta \quad \text{Surface tension} \\
\theta_{\text{c}} \quad \text{Contact angle} \\
K \quad \text{Absolute permeability}
\( D_i^0 \) Mass diffusivity of species of \( i \)
\( p_0 \) Reference pressure
\( T_0 \) Reference temperature
\( r_s \) Exponent of pore blockage
\( \gamma_p \) Exponent of pressure
\( \gamma_t \) Exponent of temperature
\( \lambda \) Water content
\( \sigma_{mem} \) Membrane conductivity
\( j_{w}^{diff} \) Back diffusion flux of water
\( \rho_m \) Density of the dry membrane
\( M_m \) Equivalent weight of the dry membrane
\( P_{wv} \) Water vapour pressure
\( I_{\text{leak}} \) Specific leakage current
LIST OF FIGURES

Figure 1.1: World total final energy consumption from 1971 to 2013 by fuel (International Energy Agency 2014)
Figure 1.2: fuel shares of CO2 emissions from fuel combustion (International Energy Agency 2014)
Figure 1.3: World electricity production from 1973 to 2013 (International Energy Agency 2014)
Figure 1.4: Schematic of a PEM fuel cell showing the main reactions (Mattuci 2001)
Figure 3.1: (a) The electrolysis of water; (b) a small current flow (Larminie and Dicks, 2003)]
Figure 3.2: Mond-Langer cell
Figure 3.3: PEM fuel cell theoretical model (Mattuci 2001)
Figure 4.1: Flowchart of the likely basic calculation method in the ANSYS Fuel Cell Module
Figure 4.2: Contour map of current density distribution in membrane
Figure 4.3: the graph of velocity of oxygen
Figure 4.4: vectors of current flux density in cross section of the fuel cell through all the layers
Figure 4.5: The dimensions of the basic fuel cell modelled with the ANSYS Fuel Cell Module
Figure 4.6: The mesh of the basic fuel cell modelled
Figure 4.7: Modelling option dialogue box in the ANSYS Fuel Cell Module
Figure 4.8: ANSYS Fuel Cell Module component models
Figure 4.9: V-I curve of a single channel fuel cell
Figure 4.10: ANSYS Fuel Cell Module parameter setting for the basic cell modelled
Figure 4.12: V-J curves for the basic PEM fuel cell obtained from the ANSYS Fuel Cell Module for various values of reference current density
Figure 4.13: V-J Curve Comparison by change of initiate water content
Figure 5.1: The design for PEM fuel cell A (Doddathimmaiah, 2008)
Figure 5.2: O-side flow-channel dimensions used in experimental PEM fuel cell A. (Doddathimmaiah, 2008)
Figure 5.3: H-side flow-channel dimensions used for the experimental Cell A. (Doddathimmaiah, 2008)
Figure 5.4: Experimental set up used by Doddathimmaiah (2008)
Figure 5.5: (a) Oxygen endplate, and (b) hydrogen endplate used in cell B
Figure 5.6: Experimental fuel cell B. The external dimensions of the cell are 7.2 cm × 8.4 cm × 9 cm.
Figure 5.7: Experimental set up for performance measurement of cell B
Figure 5.8: Measurement equipment used in the experiment with cell B
Figure 5.9: Hydrogen plate of experimental cell C
Figure 5.10: Oxygen plate of experimental cell C
Figure 5.11: The reversible fuel cell testing rig assembled in the Sustainable Energy Hydrogen Laboratory at RMIT University’s Bundoora East campus used to test cell C
Figure 5.12: 3D geometry of cell A as represented in the ANSYS module
Figure 5.13: Fuel cell flow channels in the ANSYS module
Figure 5.14: Layers and thickness of the fuel cell A as modelled
Figure 5.15: Outside and inner view of the mesh used to model the multi-channel fuel cell A in the ANSYS Module
Figure 5.16: (a) Overall view of the geometry; (b) Close view of the GDL, catalyst and membrane
Figure 5.17: Overall view of mesh (left); Close view of the GDL, catalyst and membrane (right)
Figure 5.18: V-J Curve of cell A using ANSYS Fuel Cell Module
Figure 5.19: Simulated mass fraction of hydrogen in the gas flow channel in cell A
Figure 5.20: Simulated velocity of hydrogen in the channels
Figure 5.21: Current Flux Density in the membrane of cell A (0.3V)
Figure 5.22: Current flux density magnitude in the membrane
Figure 5.23: V-J curve form ANSYS simulation result and the experimental curve
Figure 5.24: Voltage versus current density curves of experiment fuel cell B
Figure 5.25: The V-J curves obtained from the ANSYS Module simulation compared with the experimental data
Figure 5.26: Fitted ANSYS Model and experiment data
Figure 5.27: The V-J curve for fuel cell measured by experiment
Figure 5.28: Interpolation on the experimental V-J curve for cell C to obtain the experimental points corresponding to the voltage values used in the simulation
Figure 5.29: The curves of voltage vs. current density of experiment and initial input values as for cell B simulation
Figure 5.30: Simulated current density distributions for cell C (a) in hydrogen-side end plane and channel, (b) hydrogen-side gas diffusion layer, (c) hydrogen-side catalyst, (d) central plane of membrane, (e) oxygen-side catalyst, (f) oxygen-side gas diffusion layer, and (g) oxygen-side end plate and channel
Figure 5.31: Effect of cell voltage on spatial distribution of current density in the middle of the membrane.
Figure 5.32: V-J Curves from the ANSYS model compared to the experimental curve for cell C
Figure 6.1: Gas flow channel configuration. (a) hydrogen flow channel, (b) oxygen flow channel
Figure 6.2: The flow channel configuration as modelled in the ANSYS Fuel Cell Module
Figure 6.3: Overall view of the mesh used in the simulation the fuel cell
Figure 6.4: Front view of the mesh in the endplate of the fuel cell model
Figure 6.5: Mesh in gas diffusion layer, catalyst and membrane.
Figure 6.6: Current density in the membrane for the large PEM fuel cell modelled
Figure 6.7: Water content in the membrane for the large PEM fuel cell modelled
Figure 6.8: Gas flow in the hydrogen channel (a) Velocity of hydrogen (b) total pressure of hydrogen (c) Mass fraction of hydrogen (d) Molar concentration of hydrogen
Figure 6.9: Gas flow in the oxygen channel (a) Velocity of oxygen (b) Total pressure of oxygen (c) Mass fraction of oxygen (d) Molar concentration of oxygen
Figure 6.10: V-J curves of Experimental data and simulation result
Figure 6.11: V-J Curve of ANSYS simulation including liquid water formation
Figure 6.12: Volume fraction of liquid water at the boundary surface between the oxygen catalyst layer and the oxygen-side GDL at different cell voltages

Figure 6.13: Oxygen concentration at oxygen catalyst surface (touching the GDL) at different voltage

Figure 6.14: V-J curve for average current density across the whole active area, and the corresponding average water saturation – J curve at the oxygen-side catalyst layer. Note the reverse vertical scale for water saturation.

Figure 6.15: V-J curve (for average current density), and oxygen concentration versus average current density

Figure 6.16: V-J Curve and current density versus hydrogen concentration

Figure 6.17: V-J curve for the base case, higher pressure gas case, lower temperature case, compared with the fitted experiment data. The average oxygen concentrations in the catalyst layer (kmol/m3) at each point are also shown

Figure 6.18: V-J Curve for base case, higher-pressure gas, lower-temperature case, compared with the fitted experiment data with average and water saturation in the catalyst layer given at each point

Figure 6.19: Water saturation versus current density curve for base case, higher-pressure case, and lower temperature case

Figure 6.20: Oxygen concentration versus current density curve for base case, higher-pressure case, and lower temperature case

Figure 6.21: V-J curve for base case and other factor case, comparing with fitted experimental curve

Figure 7.1: configuration of the PEM fuel cell stack

Figure 7.2: ANSYS Fuel Cell Module stack management window

Figure 7.3: Oxygen channels for the stack model with gas flow direction

Figure 7.4: Geometry in the model of the four-cell stack

Figure 7.5: Overall view of the mesh in the models of stack.

Figure 7.6: Front perspective view of the mesh for stack

Figure 7.7: Maps of the distribution of current density in each cell membrane in stack

Figure 7.8: Current density distributions in the membrane of each cell

Figure 7.9: Hydrogen concentration in the hydrogen catalyst layers of each cell

Figure 7.10: 2D maps of the hydrogen concentration in the hydrogen catalyst layer of each cell in stack

Figure 7.11: Oxygen concentrations in the oxygen-side catalyst layers of each cell in stack

Figure 7.12: 2D maps of oxygen concentrations in the oxygen-side catalyst layers of each cell in stack

Figure 7.13: Gas velocity in oxygen-side channels: the overall through-hole and channels in each cell(left); and just in the oxygen-side channels of each cell (right)

Figure 7.14: Gas velocity in hydrogen-side channels: the overall through-hole and channels in each cell (left); and just in the hydrogen-side channels of each cell (right)

Figure 8.1: current density of experimental V-J curve of small single fuel cell C and large single fuel cell

Figure 8.2: current density of experiment and simulation of small single fuel cell C and large single fuel cell

Figure 8.3: Current density distribution of small single fuel cell (left) and large single fuel cell (right)

Figure 8.4: Oxygen concentration in the catalyst layer of cell C (left) and in the individual cells in the four-cell stack (right)
LIST OF TABLES

Table 3.1: Selected studies of PEM fuel cell modelling over the past ten years
Table 4.1: Input parameters required by the ANSYS Fuel Cell Module
Table 4.2: Material properties of cell components required as inputs to the ANSYS Fuel Cell Module
Table 5.1: Small single fuel cells experimented and simulated
Table 5.2: Input parameters in the ANSYS Fuel Cell Module simulation of cell A
Table 5.3: Comparison of current density of experiment and simulation for cell A
Table 5.4: The three sets of input parameters used in the simulation models of cell B
Table 5.5: Variance between experiment data and ANSYS models
Table 5.6: Current densities from the ANSYS simulations corresponding to the selected cell voltage using the three sets of input parameters, along with the respective experimental current densities
Table 5.7: Variance between experiment data and ANSYS models
Table 5.8: Major input parameter for best fitted simulation of cell C
Table 6.1: Major input parameter for simulation of large fuel cell
Table 6.2: Comparison of current density between fitted experimental curve and simulation curve
Table 6.3: The values for other input parameters that can affect the mass transfer constraint used in the base-case simulation, and changed values explored in reruns of the simulation.
Table 7.1: Input parameters for the simulation of four-cell stack
EXECUTIVE SUMMARY

Transportation is an important service in everyone's life in modern cities. On account of greenhouse gas emission and other air pollution, fossil fuel need to be replaced by other green energy sources. Hydrogen produced from renewable energy sources such as solar and wind energy, stored as a compressed gas, liquid or in the solid state, and then used in fuel cells to produce electricity, offers an attractive sustainable energy option for transport and stationary applications.

Nowadays the Proton Exchange Membrane (PEM) fuel cell is a critical component in hydrogen fuel cell vehicles and hydrogen-based energy storage systems. PEM fuel cells are thus likely to play an important role in sustainable energy economies based essentially on energy efficiency and renewable energy. The design of PEM fuel cells is a very complex multi-physics task that is greatly facilitated by a multi-component computer simulation modelling, incorporating all the necessary theoretical equations, to predict cell and stack performance. The ANSYS Fuel Cell Module is such a model that is becoming more widely used in the design and optimisation of PEM fuel cells.

The aim of this thesis is thus to apply and evaluate use of the ANSYS Fuel Cell Module in computer modelling of single PEM fuel cell and PEM fuel cell stacks, by comparison of simulation outputs with experimental results.

The key research questions addressed include:

1. To what level of accuracy can the simulations of the performance of individual PEM fuel cells using the ANSYS Fuel Cell Module match experimental performance data?
2. How does changing the membrane active area affect the performance of a single PEM fuel cell in terms of current density and power density?
3. How does the performance of a PEM fuel cell stack change compared to single fuel cells in terms of current density and power density?
4. What is the best approach for using the ANSYS Fuel Cell Module to representing the mass-transfer limited portion of the voltage-current curve of a PEM fuel cell at higher current densities?
The deliverables from this project are:

- A quantitative assessment of the validity of using the ANSYS Fuel cell Module to simulate the performance of individual PEM fuel cells
- An improved understanding of the effects of membrane active area size on performance of single PEM fuel cells,
- An improved understanding of the difference in performance between single fuel cells and fuel cell stacks on the basis of current and power densities.
- A further understanding of the mass-transfer constraint and how to use ANSYS Fuel Cell Module to represent the mass-transfer limitation in the form of Voltage-Current Curve.

Form the literature review conducted in this project, a number of gaps in knowledge and understanding relating to the simulation modelling of PEM fuel cells and stacks emerged as follows:

- There is a lack of full 3D modelling taking into account gas flows, two-phase flow regions, electrochemical reactions, and charge flow (electrons and protons) in all regions of single PEM fuel cells, with detailed comparison of the simulation results with experiments conducted on a cell of the same design and materials.
- Few papers have focussed on the effects of operating conditions – reactant flow rates, cell and gas temperatures, gas pressures, relative humidity, mass fraction of liquid water – on cell performance.
- Little work has been done to compare the performance (per unit membrane active area) of large and small single cells, with a detailed analysis of the variations in current density across the active area and the causes of these variations.
- There is a lack of understanding of empirical data on the causes of the turndown in power output at high current densities, relating to saturation of porous GDLs, gas channels, and catalyst layers with liquid water, and kinetic limitations in the electrochemical reactions.
- There has so far been relatively little experience gained in the detailed 3D modelling of PEM fuel cell stacks in which the inflows to and outflows from, and voltage of, each cell can vary.
The present thesis will seek to make a contribution to the detailed knowledge and understanding in all these areas.

During the research for this thesis, an unnecessary and unphysical limitation in the allowed input values for charge transfer coefficients in Butler-Volmer equations in the then-current version of the ANSYS Fuel Cell Module was identified. Corrections to the program script were therefore made to solve the problem. These corrections have been accepted by the ANSYS Module development team and incorporated into later versions of this software.

In this project, the ANSYS Module has been used first to simulate three small fuel cells (5 cm\(^2\) active membrane area). The ANSYS simulation output for the voltage-current density (VJ) curve of the first small fuel cell, using input parameters from an earlier study, compared closely with previous experimental results for this cell. The second small fuel cell used the same geometry of the first one, but different materials for the catalyst and gas diffusion layers. The input values for the material properties were then changed in the simulation to obtain the best fit to the experimental VJ curve obtained in the present project. A third small cell with a different flow-channel geometry was measured experimentally, and also modelled in the ANSYS Module. Varying input parameters in the simulation allowed a good fit between experimental and the simulation output for the VJ curve. The best-fit input parameter values for this cell were used in the later simulation of a larger fuel cell employing the same materials, and also in the fuel cell stack modelling.

A large fuel cell (225 cm\(^2\) active membrane area) has also been simulated using the ANSYS module, and the VJ curve obtained compared with experimental measurements for the same cell. The gas flow rates were set at about 45 times those for the small fuel cell, in proportion to the active areas. It was found that the average current density of the large cell was lower than the average current density of small fuel cell at same voltage in both the experiment and simulation. The spatial distribution of current density in the large fuel cell showed an uneven distribution of current density with large variations across the active area. The current density distribution from the ANSYS simulation was able to be explained in terms of the corresponding gas concentration distributions.

The experimental VJ curve for the large cell displayed clear indications of a sharp turndown at high current density due to mass transfer constraints. It was found that the ANSYS Module
can represent this turn-down through increasing water saturation and decreasing oxygen concentration. But it has not been possible to change the input parameter values sufficiently to get an onset of this constraint at the lower current densities found in the experimental V-J curve.

In this project, a four-cell fuel cell stack (5 cm² active membrane area for each cell) was also simulated using the ANSYS Fuel Cell Module. The four-cell fuel cell stack had through holes (one for hydrogen and the other for air/oxygen) to supply the respective gas flow channels in each cell, and further through holes to provide the corresponding gas exits. The average current density found in the simulation of the whole fuel cell stack was considerably lower than that for a comparable small single fuel cell. The current density distribution also varied from cell to cell along the stack. Tentative explanations of these variations are offered.

Overall this thesis has concluded that the ANSYS Fuel Cell Module is a very useful tool for the simulation of PEM fuel cells, particularly in the lower current density range. Simulation modelling is virtually the only way to study the spatial variation of key parameters such as current density and water saturation across the active area of a cell. A great deal of further research, however, is needed in accurate representation of the mass transfer constraint in PEM fuel cells, and in modelling fuel cell stacks.

The following recommendations are made relating to the further development of simulation of PEM fuel cell:

- More research needs to be done on the relationship between gas flow rates and the gas concentrations in the catalyst layers, and hence the effects on current density in single PEM fuel cells, and for each cell within a stack.
- Further study needs to be done on the influence on PEM cell performance of flow channel width and distance between channels by varying these dimensions while keeping the gas concentration constant.
- There needs to be more research conducted into what input parameters can be changed, or actual changes made to the basic code, in order for the ANSYS Fuel Cell Module to represent more accurately the onset and magnitude of the mass transfer constraint, and its relation to water saturation in the VI curves for single PEM fuel cells.
- Further systematic research using ANSYS Fuel Cell Module simulations should be conducted into evaluating the relative effects on performance of increasing the active
membrane area per cell, versus increasing the number of cells of a given area in a stack, while keeping the increase in total active area constant, with validation of the findings against experiments.

- Further experience should be gained in the use of computer simulation modelling of PEM fuel cell stacks, by the ANSYS Module or other software, to optimise design features such as:
  - flow channel configuration on both hydrogen and oxygen sides
  - oxygen and hydrogen GDL material and geometric properties
  - catalyst properties such as specific surface area (that is, active area per unit volume).
1. INTRODUCTION

1.1 BACKGROUND

The energy supply of society is currently based mainly on fossil fuels. The total world final consumption of energy from 1971 to 2013 is shown in Figure 1.1.

Since 1971 all forms of energy consumption have been increasing, with oil consumption always taking the largest part. Total final consumption of oil in 2013 was 39.9% of total consumption. The fossil fuel including oil, coal and natural gas accounted for 66.5% of world total final consumption.

Furthermore, although electricity took 18% of world total final consumption, most electricity was generated using fossil fuels. Electricity generated by coal also reached the highest level at 9614 TWh in 2013, about 41.1% of global electricity production (International Energy Agency 2014). While electricity was mostly generated by coal and natural gas, accounting for over 60% of total electricity in 2013 (International Energy Agency 2014), the total consumption of fossil fuels was even more than 66.5%.

Many reports have projected that many critical forms of fossil fuel will be used up soon (Shafiee & Topal 2009; Rahman et al. 2014). Especially oil is a most critical one, since people’s normal life is based so heavily on oil. As the U.S. Energy Information Administration (2014) reported, proven reserves of crude oil in 2014 were 1,655 Billion
Barrels. The production of crude oil is 93 018 thousand barrels per day, so at this rate of production the oil would be used up in just half a century. It is therefore quite urgent to develop other kinds of energy source, which preferably should be renewable and have as near equivalent energy density as possible, to replace oil in many areas as energy supply, mainly in transport.

![Chart showing fuel shares of CO₂ emissions from fuel combustion](image)

Figure 1.2: fuel shares of CO₂ emissions from fuel combustion (International Energy Agency 2014)

Over and above the limited and finite supply of fossil fuels, the greenhouse gas emissions, mainly carbon dioxide, from their combustion has emerged as a major constraint on their continued use over the past few decades. The world-wide climate has changed a lot in recent years in an alarming fashion (IPCC 2014) due to rising atmospheric level of greenhouse gases, particularly carbon dioxide from burning fossil fuels. In 2013, the carbon dioxide emission was 32 190 Mt/y, and over 99% was from fossil fuels (Figure 1.2).
As shown in Figure 1.3, renewable electricity generation had become the second largest source of electricity generation worldwide by 2014, accounting for 22% of total electricity. But coal was still the largest electricity generation source, about twice that of renewables. Coal combustion is one of main sources of carbon dioxides emission. As shown in Figure 1.2, about 46% of CO₂ emission is from coal combustion. Hence in order to reduce carbon dioxide emissions, an environmental friendly energy source should be used to replace fossil fuel. Nowadays, renewable energy sources like wind energy and solar energy are becoming widely used for electricity generation. But wind energy and solar energy are not constant energy sources, and cannot provide continuous and reliable electricity, so these energy sources cannot replace the coal only depend on themselves. However, with energy storage, renewable energy can provide reliable constant power supply.

Currently batteries are most widely used electricity storage, especially lithium ion batteries. Another alternative is to use a hydrogen-based system for energy storage with renewables, where surplus electricity over demand can be used to generate hydrogen by water splitting in an electrolyser. Hydrogen gas can be stored by various means, and reused subsequently in fuel cell to supply electricity when there is insufficient primary renewable energy to meet demand (Shabani & Andrews 2011; Fathabadi 2017).

A hydrogen fuel cell was first demonstrated in 1839 by William Grove (Larmine & Dicks 2003). In the reverse reaction to electrolysis, hydrogen and oxygen are electrochemically recombined in a fuel cell, without combustion of the hydrogen, to produce an electric current.
Such a fuel cell is environmentally friendly, since the only reaction product is water (Larminie & Dicks 2003). Hydrogen fuel cells can also provide a viable alternative to batteries as a power source for electric vehicles (Ceja 2011; Toyota 2017). Hydrogen is normally produced by electrolysis of water. If the electricity for electrolysis of water is produced from renewable energy, then the hydrogen fuel cell vehicle is absolutely zero greenhouse gas emission. Fuel cell vehicles promise to cost less to run over the same distance than battery electric vehicles (Nancy et.al 2012), since the cost of producing the hydrogen for the fuel cell is generally less than that of charging a lithium battery (Blencoe 2009). A hydrogen fuel system should have a much longer lifetime than a comparable battery (Battery University, 2011; Department of Energy USA, 2008). Fuel cell transportation systems can provide a higher system specific power at 420 W/kg, (Steven & James, 2006) while lithium batteries have a specific power of 350 W/kg. (Kahligh & Li 2010; Stempien & Chan 2017).

Andrews and Shabani (2012) have argued that hydrogen has a critical role to play in a sustainable energy economy based essentially on energy efficiency and renewable, serving as the energy storage needed to guarantee continuous supply, and as transportable fuel to replace petrol and diesel. Hydrogen has the highest energy to mass ratio of all fuels. 1 kg of hydrogen contains the same amount of energy as 2.1 kg of natural gas or 2.8 kg of gasoline (Santhanam 2016). On the other hand, hydrogen gas has a very low volume energy density at normal pressures. The storage of hydrogen is thus a major problem that has to be solved before the hydrogen economy can begin to be implemented on a large scale. Nowadays, the common ways to store hydrogen are as compressed gas, liquid hydrogen, and in the solid state in metal hydrides (Department of Energy USA 2008). If hydrogen is carried and used on board a vehicle, clearly safety considerations are very important. On ignition hydrogen burns in air at volumetric hydrogen concentrations from 4 to 74.5 %. The new solid-state hydrogen storage technology of metal hydrides is an attractive way to solve the safety problem, and to achieve competitive levels of energy storage per unit mass and volume (Andrews & Shabani 2012), although high-pressure compressed gas storage in lightweight advanced composite thanks has currently achieved the highest gravimetric energy density, such as Toyota’s Mirai fuel cell vehicle with 5.7 wt% for hydrogen stored at 700 bar (Toyota 2017).
1.2 Research on Proton Exchange Membrane Fuel Cell

A lot of research has been done on various aspects of PEM fuel cells. The standard single PEM fuel cell is a combination of two endplates as current collectors, two gas diffusion layers, two catalyst layers and a proton exchange membrane (Figure 1.4).

![Image of PEM fuel cell](Figure 1.4: Schematic of a PEM fuel cell showing the main reactions (Mattuci 2001))

Generally, the hydrogen is fed in from anode side channel and split in the catalyst layer into protons and electrons. The protons pass through the membrane to the cathode catalyst where they combine with the oxygen fed in from the cathode-side channel and electrons from the external electric circuit to form water. The movement of the electrons in the external circuit is the current generated.

The most commonly studied areas are the best materials for the membrane, catalyst and gas diffusion layers, the flow channel design, and the cooling channel design. Metha and Copper (2003) reviewed 16 polymer electrolyte membranes, two types of gas diffusion layers, eight types of anode catalysts, four types of cathode catalysts and over 100 bipolar plate designs in their review paper. This work not only reviewed membrane electrode assembly manufacturing options and synthesis processes for many of the membranes and for the gas diffusion layers, but also covered the bipolar plate fabrication options. The advantages, challenges and progress of various cooling technique – including heat spreaders, separated air flow, liquid cooling, and cooling with a phase change materials – have been reviewed by
Zhang & Kandlikar (2012). Effective cooling is critical for safe and efficient operation of proton exchange membrane fuel cells with high power.

With respect to shape factors such as exterior of fuel cell, some papers have investigated cylindrical PEM fuel cells (Bullecks et al. 2011, Maher 2008), comparing the performances of cylindrical and planar cells using the same MEA. The results showed that the cylindrical cell performed better than the planar cell in the higher overpotential regions. Hence the further investigation of cylindrical PEM fuel cells is of research interest. More generally, there has been little work done to date that has systematically studied the effects of varying the shape of the fuel cell – both that of the active membrane area, and the number of cells in a stack – on cell and stack performance. The present project – will therefore investigate the effects of variation in the active membrane area and the flow-channel configuration, as well as stacking multiple cells in series, on overall performance.

Because of the spatially-enclosed nature of a PEMFC, it is not easily possible to measure internal quantities experimentally. The use of computational fluid dynamics (CFD) is an attractive option for analysis of species concentrations, temperature gradients or pressure distributions throughout each component of a PEMFC, and the effects of cell geometry on these parameters. A number of papers have discussed conservation equations in general and PEMFC-related micro- and macroscopic heat and mass transfer in particular. For example, Siegel (2008) focussed on published CFD models and additionally captures some computational aspects and solution procedures. As for the software, FLUENT, COMSOL Multiphysics (FEMLAB), STAR-CD, CFD-ACE+, and Open FOAM have all been used for PEMFC modelling. Siegel (2008) introduced these software packages and some case studies on their use in analysing a range of PEMFC-related topics. FLUENT is one of the most complete CFD software available and comes with powerful pre- and postprocessors. Finite volume computations can be performed in single or double precision using different solvers.

ANSYS FLUENT simulation tools are capable of modelling and predicting the electrical, thermal and fluid performance of a single fuel cell — as well as complex fuel cell stacks (ANSYS 2012). Powerful computational fluid dynamics tools from ANSYS can help to streamline both cell- and stack-level testing, using numerical modelling to arrive at optimised fuel cell designs with superior power generation rates, reliability and durability. Simulation
tools from ANSYS also help in assessing the role of thermal and electromechanical stresses on cell and stack performance. Hence the ANSYS Fuel Cell Module, which combines the CFD, electrostatic, and thermal simulation tools in the ANSYS suite, with a specific electrochemical model of a number of types of fuel cell, including PEM fuel cells, has been judged to be the most suitable for application in the present study.

1.3 AIM AND OBJECTIVES OF THIS THESIS

The aim of this thesis is thus to apply and evaluate the use of the ANSYS Fuel Cell Module in computer simulation modelling of single PEM fuel cells and PEM fuel cell stacks, with comparison of simulation outputs with experimental results.

The primary objectives of this research are to:

1. Test the validity of the ANSYS Fuel Cell Module against experimental data for simulating the performance of single PEM fuel cells
2. Gain an improved understanding of the effects of size of membrane active area on the performance of PEM fuel cells using both ANSYS computer simulation modelling and experiments
3. Apply the ANSYS Fuel Cell Module to a small PEM fuel cell stack and compare the performance to that of a single PEM cell of the same active membrane area as each cell in the stack
4. Study how effectively this Module represents the effects of mass-transfer limitations on the voltage – current curve of PEM fuel cells at high current densities.

1.4 RESEARCH QUESTIONS

The key research questions addressed are:
1. To what level of accuracy can the simulations on the performance of individual PEM fuel cells using the ANSYS Fuel Cell Module match experimental performance data?

2. How does changing the membrane active area affect the performance of a single PEM fuel cell in terms of current density and power density?

3. How does the performance of a PEM fuel cell stack change compared to single fuel cells in terms of current density and power density?

4. What is the best approach for using the ANSYS Fuel Cell Module to represent the mass-transfer limited portion of the voltage current density curve of a PEM fuel cell at higher current densities?

1.5 SCOPE

The thesis will mainly focus on modelling and simulation of PEM fuel cell, not other types of fuel cell.

Because this project has contributed to a major RMIT University research, development and demonstration project investigating reversible PEM fuel cells (specifically Unitised Regenerative Fuel cells; and to the RMIT CTD Project), some of the experimental results on fuel cell performance are for reversible fuel cells operating in fuel cell mode. No simulations are reported in this thesis for these URFCs running in electrolyser mode.

Other key limitations in scope are the following:

- The ANSYS Fuel Cell Module has been exclusively used to run the simulations
- Rectangular header-riser gas flow channel configurations are used; with the large single fuel cells modelled the effect of partitioning the overall active area into four separately supplied quadrants is investigated.
- The main variation for single fuel cells investigated is the size of the membrane active area (5 cm$^2$ and 225 cm$^2$) and its influence on current and power density (that is, current and power per unit active area).
- The small PEM fuel cell stack system considered had four cells, with the membrane active area the same as used the with small single fuel cell modelling, namely 5 cm$^2$. 
In the simulation modelling, the fuel cell stacks are supplied with gas to each layer via a common through-channel with connections from this to the inlet of flow-channel array in each layer.

Hence, while this study has significantly advanced the practice and understanding of PEM fuel cell computer modelling, what could be achieved within the time and resource constraints of a single PhD thesis has inevitably been limited. Thus it remains a preliminary investigation into the vast and highly complex field of integrated PEM fuel cell simulation modelling, with the influence of numerous design factors on PEM fuel cell and stack performance still to be systematically studied in the future.

1.6 DELIVERABLES

The deliverables from this project are:

• A quantitative assessment of the validity of using the ANSYS Fuel cell Module to simulate the performance of individual PEM fuel cells
• An improved understanding of the effects of membrane active area size on the performance of single PEM fuel cells,
• An improved understanding of the difference in performance between single fuel cells and fuel cell stacks on the basis of current and power densities.
• A further understanding of mass-transfer limitation and how to use ANSYS Fuel Cell Module to represent the mass-transfer limitation in the form of voltage versus current density curve.

1.7 GUIDE TO THESIS

The overall thesis is organised as follows.

Chapter 2 explains the research method and plan, including the methodological approach and each main phases and activities of the research project.
Chapter 3 begins with a brief history of the development and application of PEM fuel cells, and their current state of the art. It then presents a review of proton exchange membrane (PEM) fuel cell simulation modelling and its applications to date, including comparisons with experimental data on fuel cell performance. The various computer simulation models available are presented and briefly compared. The chapter concludes by identifying gaps in knowledge and understanding in this area of scientific and technological study, which have guided the definition and conduct of the present research project.

The computer simulation tool used in the project is the ANSYS Fuel Cell Module. An introduction to this Module is provided in chapter 4. The required inputs to and available outputs from this module are explained. Also in this chapter, an important correction to the script of the Butler-Volmer equations used in ANSYS Fuel Cell Module (version 12.0) that has been discovered and worked out in the course of the present thesis project is reported. This correction relates to the allowed inputs of the charge exchange coefficients on anode and cathode side. A basic PEM fuel cell with a single pair of gas flow channels (one for hydrogen, and the other for oxygen) is then modelled using ANSYS Fuel Cell Module to illustrate how the Module can be used to investigate the effects of varying inputs on outputs and cell performance.

The simulation of small single PEM fuel cells with the ANSYS Fuel Cell Module is presented in chapter 5. The results of the simulations are compared with three experimental fuel cells, and input parameters varied to achieve the best fit to the experiment polarisation (voltage-current) curve. Values for these input parameters to gain the most accurate representation of the output of real fuel cells are thus obtained. These input parameters cannot be measured by common physical methods. The values obtained were then used subsequently in the ANSYS simulation modelling of large single fuel cells and fuel cell stacks.

A simulation of a large single PEM fuel cell is present in chapter 6. The experimental data obtained are compared with the simulation results. Adjustments to the ANSYS model of the large single PEM fuel cell are made to match more closely to the experimental data. In addition, in this chapter, the way the Module represents the effects of mass-transfer limitations on the voltage-current curve of PEM fuel cells at high current densities are investigated in detail. By applying these ways to a simulation of a single large cell operating
at high current density, the limitations of the ANSYS Module in representing saturation or mass-transfer limits are studied, and some options to improve the Module in this research are identified.

As individual fuel cells must be arranged in multi-cell stacks to provide significant power in any application, chapter 7 focusses on simulation modelling of fuel cell stacks in the ANSYS Fuel Cell Module. In this chapter, the difference between a single fuel cell and fuel cell stacks is studied, and an understanding of the effect of gas distribution on the current density distribution is developed.

Finally, chapter 8 presents the answers to each of the research questions posed at the beginning of the thesis, and then draws a number of other conclusions. Recommendations for further work in this area are made.

Published conference paper is listed in the end of the thesis. More journal papers arising for this thesis are prepared and going to be submitted.
2. METHOD

2.1 OVERALL METHODOLOGICAL APPROACH

2.1.1 Computer simulation modelling

The design for PEM fuel cell is complex, since many input parameters influence the performance of the cell, including: the flow channel configuration, the size of active membrane area, the flow channel geometry, the thickness of each component, and the material of gas diffusion layers, catalysts and membranes. In addition, the main physical and electrochemical processes taking place – such as diffusion of input gases through gas flow channels, gas diffusion backing layers to the catalyst layers deposited on the Nafion membranes, the electrochemical reaction that take place on the catalysts, the conductivity of protons (in the form of hydronium) through the Nafion membrane, the formation and removal of product water on the oxygen side electrode, and the conduction of electrons through endplates and bipolar plates, and gas diffusion backing layers – all require many equations to describe theoretically. These equations usually can only be solved using numerical methods. Hence it is essential to have a multi-component computer simulation model, incorporating all the necessary theoretical equations, to model PEM cell and stack performance.

Since lots of variable parameters need to be considered when developing PEM fuel cell designs, the effect of each parameter should be studied in order to develop the most suitable design to meet the demand. It would be prohibitively time-consuming and expensive to use experiments to test every possible design variation. Hence using numerical computer simulation models to simulate the performance of the fuel cell under a variety of input assumptions is of great benefit, and much more efficient in terms of time and costs.

The first computer simulations conducted were based on simple one-dimensional numerical fuel cell models, and then later computer simulations began to use three-dimensional models. The present project is aiming to contribute to complete PEM fuel cell and stack design, so
that PEM fuel cells of different sizes, different gas flow management and different operating temperatures need to be simulated. The outputs of the simulations are required to include not only the power performance, current, voltage, but also current distribution, gas distribution, water formation, pressure change, gas flow as well. Fortunately, such a model has already been developed in the form of the ANSYS Fuel Cell Module, which incorporates finite-element programs (ANSYS Fluent) for computational fluid dynamics to model fluid flow in both liquid and vapour phases, and an electromagnetic suite of programs to model electrical conductivity, both electron and proton, through various materials. In addition, the ANSYS Fuel Cell Module uses the standard Butler-Volmer equations to represent the electrochemical reactions on the electrodes. Hence this Module has been used in the present project to investigate the effects on PEM fuel cell performance of flow channel configuration and geometry, the size of membrane active area and change from single fuel cell to fuel cell stack. This module is useful for the evaluation of designs of PEM fuel cells and stacks. It also can provide the graphical plots of the current distribution in any plane that can be defined through the geometry of the fuel cell. In order to get the best output performance, an even distribution of current density across the membrane active area is important to avoid any hot spot which will limit the total performance. Further discussion on the choice of the ANSYS Fuel Cell Module for the present study, rather than other models available for these tasks, is provided in chapter 4.

2.1.2 Validating the ANSYS Fuel Cell Module against experiment

However, it is necessary to validate the outputs of the computer simulation modelling against experimental results in a number of specific cases before applying the model more generally. In the present thesis, the simulation cases used were as follows:

- Cell A, Arun Kumar Doddathimmaiah’s cell constructed at RMIT in 2008 (Doddathimmaiah 2008)
- Fuel cell B and fuel cell C constructed for this project with 5 cm² membrane active area, operating with pure oxygen for Cell B and air for cell C.
- A larger individual fuel cell with membrane active area of 225 cm² designed for later incorporation into a fuel cell stack
• A fuel cell stack comprised of four cells each with the same membrane active area as the single fuel cell C (5 cm²), with gas gas fed to the individual cells in the stack via through-holes passing right through all plates from an end plate.

Fuel cell A, B and C are used in validating the ANSYS Fuel Cell Module, since these have also been constructed and tested as experimental cells with measured data to compare with simulation results. An earlier PhD student Arun Kumar Doddathimmaiah in SAMME had made a small fuel cell and measured its performance experimentally (Doddathimmaiah 2008). The results obtained from this earlier work were used to calculate the parameters that were not able to be directly measured experimentally, such as reference exchange current densities and charge exchange coefficients.

The large number of finite elements in the simulation models required a long time for calculation. Time and constraints on access to high-performance computers led to the decision to restrict the scope of the study to a limited number of small fuel cells, plus just one larger cell. The experimental fuel cells had 5 cm² active membrane area with parallel flow channels on both sides.

Simulation models are used to explore the influence of major design parameters on performance. There are results for three experimental fuel cells that are compared with simulation results using ANSYS Fuel Cell Module. The first fuel cell A had been measured previously by experiment, and the input settings for the ANSYS Fuel Cell Module simulation were taken mainly from the previous experimental report. Hence it was expected that the output of the simulation would be close to the experimental data, which it turned out to be. The second case is fuel cell B. The experimental fuel cell B was built especially for this project as a new cell. The geometry of cell B was the same as for cell A, while the gas diffusion layer, catalyst and membrane were newly bought from commercial producers. Hence the properties of these material may have been different from those in the previous experimental cell (A). In the case of cell B therefore the actual experimental performance results, such as Voltage vs. Current Density (V-J) curve, were compared with the simulation results from ANSYS fuel cell module. Some of the input parameters, such as reference current density, exchange coefficient, were changed to m to match the simulation output to the experiment result. The comparison was made not only by visual examination, but also by
2.1.3 Applying ANSYS module to larger single fuel cell and fuel cell stack

The ANSYS module was used to simulate the small single fuel cells, and a set of input parameters was obtained that gave the best-fit of ANSYS simulation to the experimental result. This set of input parameters was then used in the simulation of a single large fuel cell to investigate the effect of the size of membrane active area on the cell performance.

This set of input parameters was also used in the simulation of a small fuel cell stack to compare its performance to a single fuel cell. The small fuel cell stack was a four-cell fuel cell stack with 5 cm² membrane in each cell, and this was compared with the single fuel cell C (also 5 cm²) in terms of current density and power density.

2.1.4 Limitations of this method

There is always a risk that by varying input parameters in a simulation model to get the best fit to experimental performance that the ‘validation’ of the model is assured, by essentially a circular process. Care was taken to avoid this circularity by always checking the best-fit values obtained were in the ranges indicated by previous studies that had been compared with experiment. Also the best-fit values obtained from a comparison with experiment for a particular cell were then tested by applying to a cell with some variations in design (for example, active area) but employing the same materials (in catalysts and gas diffusion layers, for example); then this new simulation result was compared to a separate experiment for the different cell.
Whenever key input parameters to a simulation cannot be directly and independently measured, however, it is always important to remain very aware of the limitations of the modelling exercise.

2.2 MAIN PHASES AND ACTIVITIES

2.2.1 Literature review

The literature review in chapter 3 starts with the recent state of renewable energy and the development of the PEM fuel cell. Then the review focus on the available computer simulation models and use of computer simulation on study of the effect of shape factors and gas flow channel configuration. In the end, gaps in knowledge and understanding in the area of computer simulation modelling of PEM fuel cells are identified.

2.2.2 Application of the ANSYS Fuel Cell Module to a basic fuel cell with a single straight channel for each gas

A basic PEM fuel cell with a single straight channel for each gas is simulated in ANSYS Fuel Cell Module to illustrate how the ANSYS Fuel Cell Module works in chapter 4. The basic fuel cell has an active area in the shape of a long rectangle or strip with a single straight gas flow channel on each side of the MEA, one for hydrogen and the other for oxygen. The input parameters required by the ANSYS Fuel Cell Module are identified. The effects of selected input parameters, such as reference current density and exchange coefficient, on the performance of simulation model are studied.
2.2.3 Comparison of ANSYS Fuel Cell simulation modelling of small single fuel cells with experimental data

A model of a small-scale PEM fuel cell is developed in the ANSYS Fuel Cell Module, and simulations run with various sets of input parameters. The simulations outputs are compared with experimental data for three PEM fuel cells with the same design as that simulated. The voltage-versus-current density curve obtained from the ANSYS Fuel Cell Module is compared to the corresponding experimental curve to find out to what extent can the ANSYS Fuel Cell Module represent the performance of real cells. One set of input parameters representing material properties is selected for use in modelling further fuel cell employing similar materials later in the thesis, such as a single large fuel cell and small fuel cell stacks.

2.2.4 Investigation of the effects of membrane active area on a single large fuel cell performance

Sets of models focussing on different factors, such as the shape and size of the active membrane area, are used to study the effect of each factor on the performance of fuel cell. Experimental tests of some of the design of these fuel cell designs are compared with simulation results.

A single large fuel cell is developed to study the effect of size of active membrane area on the performance of fuel cell. The input values which are not measurable, such as reference current density and exchange coefficient, are obtained from the simulation of small single fuel cells.

2.2.5 Investigation of the Mass-transfer constrain in ANSYS Fuel Cell Module

The mass-transfer constrain is studied using ANSYS Fuel Cell Module comparing with experimental result. The simulation work is done with the large fuel cell model, changing different values of temperature, pressure and other factors, to investigate the effect on the mass transfer constrain.
2.2.6 Investigation of the difference between performance on fuel cell stacks and performance on single fuel cells

The performance of fuel cell stacks and single fuel cells with comparable active area are compared using the ANSYS Fuel Cell Module. The simulation work is on a four-cell fuel cell stack with separate gas inlet for each layer, and with through channels on anode sides and cathode sides connecting the inlets or outlets on each side together.

2.2.7 Drawing conclusion and making recommendations

Finally, conclusions based on all the experimental and simulation work are drawn, and recommendations made for further investigations.
3. REVIEW OF PEM FUEL CELL

HISTORY, SIMULATION MODELLING

AND ITS EXPERIMENTAL VALIDATION

3.1 PROTON EXCHANGE MEMBRANE FUEL CELLS

3.1.1 Brief history of hydrogen fuel cells

The first demonstration of a fuel cell was by lawyer and scientist William Grove in 1839, (Larminie and Dicks 2003) using an experiment along the lines of that shown in Figure 3.1. In Figure 3.1a, water is being split by electrolysis into hydrogen and oxygen by passing an electric current through it. In Figure 3.1b, the power supply has been replaced with an ammeter, and a small current is flowing. The electrolysis is being reversed. The hydrogen and oxygen are recombining, and an electric current is being produced. Electrical energy is produced, and also some heat, from the energy liberated by the water formation reaction (Larminie and Dicks 2003).
Grove (1843) conducted a series of experiments with what he termed a gas voltaic battery, which ultimately proved that electric current could be produced by an electrochemical reaction between hydrogen and oxygen over a platinum catalyst. Ludwig Mond with his assistant Dr Charles Langer, tackled the problem of converting the Grove gas battery into something with a useful power output. (Bacon 1969) The problem was to get a sufficiently large area of platinum, both dry enough for the hydrogen to be able to get at it, and wet enough for the necessary ionic currents to flow in the electrolyte. Their solution was to trap the electrolyte in a porous matrix, such as unglazed earthenware. Against this, they pressed electrodes of thin platinum or gold foil, perforated all over with fine holes. Then they brushed on platinum black, which acted as the catalyst. With this cell (Figure 3.2) they achieved a conversion efficiency, from the total heat of reaction to electrical energy, of about 50 percent, working with pure hydrogen at room temperature (Bacon 1969).

Figure 3.1: (a) The electrolysis of water; (b) a small current flow (Larminie and Dicks, 2003)

Figure 3.2: Mond-Langer cell
In 1932, Cambridge engineering professor Francis Bacon modified Mond and Langer’s equipment to develop the first alkaline fuel cell (AFC) (Bidault and Middleton 2012), but it was not until 1959 that Bacon demonstrated a practical 5 kW fuel cell system. At around the same time, Harry Karl Ihrig fitted a modified 15 kW Bacon cell to an Allis-Chalmers agricultural tractor. (Lucia 2012) Allis-Chalmers, in partnership with the US Air Force, subsequently developed a number of fuel cell powered vehicles including a forklift truck, a golf cart and a submersible vessel.

In the late 1950s and early 1960s NASA, in collaboration with industrial partners, began developing fuel cell generators for a manned space mission (NASA 2010). The first PEMFC unit was one result of this, with Willard Thomas Grubb at General Electric (GE) credited with the invention (Lucia 2012). Another GE researcher, Leonard Niedrach, refined Grubb’s PEMFC by using platinum as a catalyst on the membranes. The Grubb-Niedrach fuel cell was further developed in cooperation with NASA, and was used in the Gemini space programme of the mid-1960s (Lucia 2012). International Fuel Cells (IFC later UTC power) developed a 1.5kW AFC for use in the Apollo space missions (UTC 2010). The fuel cell provided electrical power as well as drinking water for the astronauts for the duration of their mission. IFC subsequently developed a 12 kw AFC, used to provide on board power on all space shuttle flights.

The 1970s saw the emergence of increasing environmental awareness amongst governments, businesses, and individuals (Lucia 2012). Prompted by concerns over air pollution, clean air legislation was passed in the United States and Europe (EPA 2013). This ultimately mandated the reduction of harmful vehicle exhaust gases and was eventually adopted in many countries around the world. The 1970s was also the era of the OPEC oil embargoes, which led governments, businesses and consumers to embrace the concept of energy efficiency (U.S. Department of State 2013). Clean air and energy efficiency were to become two of the principal drivers for fuel cell adoption in subsequent decades, in addition to the more recent concerns about climate change and energy security. In 1965, General Motors had experimented with its hydrogen fuel cell powered Electrovan fitted with a Union Carbide fuel cell (Bowman 1966). Although the project was limited to demonstrations, it marked one of the earliest road-going fuel cell electric vehicles (FCEVs). From the mid-1960s, Shell was
involved with developing direct methanol fuel cell (DMFC), where the use of liquid fuel was considered to be a significant advantage for vehicle applications. Concerns over oil availability in the 1970s led to the development of some one-off demonstration fuel cell vehicles, including models powered by hydrogen or ammonia, as well as of hydrogen-fuelled internal combustion engines (Verhelst and Wallner 2009). Several German, Japanese and US vehicle manufacturers and their partners began to experiment with FCEV in the 1970s, increasing the power density of PEMFC stacks and developing hydrogen fuel storage systems. By the end of the century, all the world’s major carmakers had active FCEV demonstration fleets as a result of these early efforts. The focus by then had shifted back to a pure hydrogen fuel, which generates zero harmful tailpipe emissions (Polle 2014). Prompted by concerns over energy shortage and higher oil prices, many national governments and large companies initiated research projects to develop more efficient forms of energy generation in the 1970s. (U.S. Department of State 2013) One result of this was important advances in phosphoric acid fuel cell (PAFC) technology, in particular in stability and performance. There were significant field demonstrations of large stationary PAFC units for prime, off-grid power in the 1970s, including a 1MW unit developed by IFC. (Behling 2012) Funding from the US military and electrical utilities enabled developments in molten carbonate fuel cell (MCFC) technology, such as the internal reforming of natural gas to hydrogen. The use of an established natural gas infrastructure was a key advantage in developing fuel cells for large stationary prime power applications (Barelli et al. 2016).

Substantial technical and commercial development continued in the 1980s, notably in the area of PAFCs (Behling 2012). A bright future for the technology was widely predicted around this time for stationary applications and buses (FuelCellToday 2011). Ambitious conceptual designs were published for municipal utility power plant applications of up to 100MW output. Predictions of tens of thousands of large stationary PAFC plants in operation by the end of the century were made, but saw little commercial traction in the 1980s. With subsequent advancements in membrane durability and system performance, PAFCs were rolled out in greater numbers almost two decades later for large-scale combined heat and power application. (UTC Power 2014). Also in 1980s, research, development and demonstration work continued in to the use of fuel cells for transport applications. The US Navy commissioned studies into the use of fuel cells in submarines where highly efficient, zero-emission, near-silent running offered considerable operational advantages
In 1983 the Canadian company Ballard began research into fuel cells, and was to become a major player in the manufacture of stacks and systems for stationary and transport applications in later years (Ballard, 2013).

Attention turned to proton exchange membrane fuel cell (PEMFC) and solid oxide fuel cell (SOFC) technology in the 1990s, particularly for small stationary applications (FuelCellToday 2011). These were seen as offering a more imminent commercial potential, due to the lower cost per unit and a greater number of potential markets – for example backup power for telecoms sites and residential micro-CHP. In Japan (Carter 2012), Germany (Santarelli and Torino 2014) and the UK, there began to be significant government funding devoted to developing PEMFC and SOFC technology for residential micro-CHP applications. Government policies to promote clean transport also helped drive the development of PEMFC for automotive applications (U.S. Department of State 2013). In 1990, the California Air Resources Board introduced the Zero Emission Vehicle Mandate (Shaheen, Wright and Sperling 2002). This was the first vehicle emissions standard in the world predicated not on improvements to the internal combustion engine but on the use of alternative powertrains. Carmakers such as the-DaimlerChrysler, General Motors, and Toyota, all of which had substantial sales in the U.S., responded to this by investing in PEMFC research (FuelCellToday 2011). Companies other than automakers, such as Ballard, continued PEMFC research for automotive and stationary clean power. Ballard went on to supply PEMFC units to Daimler and Ford (Ballard 2013). The programs initiated in the 1990s still continue, albeit with some changes to the strategic focus of some key players. Significant advances in DMFC technology occurred around the same time, as PEMFC technology was adapted for direct methanol portable devices. Early applications included portable soldier-borne power and power for devices such as laptops and mobile phones. MCFC technology, first developed in the 1950s, made substantial commercial advances in the 1990s, in particular for large stationary applications in which it was sold by companies such as Fuel Cell Energy and MTU. SOFC technology also underwent substantial developments in terms of power density and durability for stationary applications. Boosted by general optimism in high-technology industries, many fuel cell companies listed on stock exchanges in the late 1990s, only for prices to fall victim to the crash in technology stocks shortly after (FuelCellToday 2011).
The European Union, Canada, Japan, South Korea, and the United States are all engaged in high-profile demonstration projects, primarily of stationary and transport fuel cells and their associated fuelling infrastructure (Kamarudin, Achmad and Daud 2009). The genuine benefits that fuel cell technology offers over conventional technology have played a part in promoting adoption. For example, the value proposition that fuel cell materials handling vehicles offer regarding extended run-time, greater efficiency and simplified refuelling infrastructure compared with their battery counterparts makes them attractive to warehouse operators. (FuelCellToday 2011) Tens of fuel cell buses were developed in the mid-2000s as part of the HyFleet: CUTE project in Europe, China and Australia. (Ruete 2009). Buses were, and still are, seen as a promising early market application of fuel cells due to their combination of high efficiency, zero-emissions and ease of refuelling, and due to the vehicles running on set routes and being regularly refuelled with hydrogen at their bases (Hoffmann 2012).

Fuel cells began to become commercial in a variety of applications in 2007, when they started to be sold to end-users with written warranties and service capability, and met the codes and standards of the markets in which they were sold (Fuel Cell Today 2011). As such, a number of market segments became demand driven, rather than being characterized by oversupply and overcapacity. In particular, thousands of PEMFC and DMFC auxiliary power units were commercialised in leisure applications, such as boats and campervans, with similarly large numbers of micro fuel cell units being sold in the portable sector in toys and educational kits (U.S. Department of Energy 2014). Demand from the military also saw hundreds of DMFC and PEMFC portable power units put into service for infantry soldiers, where they provided power to communications and surveillance equipment and reduced the burden on the dismounted soldier of carrying heavy battery packs. A large-scale residential CHP programme in Japan helped stimulate commercial stationary PEMFC shipments. These units began to be installed in homes from 2009, and more than 13,000 such units have been installed to date (Carter 2012). Demonstration programs for backup power systems in the USA gave further impetus to the stationary sector. This was also driven by practical concerns over the need for reliable backup power for telecoms networks during emergencies and rescue operations (FuelCellToday 2011). The inadequacy of diesel generators was illustrated during the Gulf of Mexico Hurricane Katrina disaster (Waple 2005), when many ran out of fuel, disrupting the telecoms network and hampering relief efforts. The need for reliable on-
grid or off-grid stationary power in developing countries also gave a boost to fuel cells. In the late 2000s, hydrogen and natural gas fuelled PEMFC units began to be sold in parts of India and east Africa to provide primary or backup power to mobile phone masts (FuelCellToday 2011). The rapidity of mobile phone adoption in these regions means that the conventional grid infrastructure cannot keep pace with new power demands, or is to unreliable for an effective mobile network (FuelCellToday 2011). Fuel cells provide a solution to this previously unmet need. In transport applications, the greatest commercial activity occurred in the materials handling segment, where there is a strong business case for their use in place of the incumbent technology, lead acid batteries (Battery University 2011). Funding for demonstration fleets of fuel cell materials handling vehicles saw increasing numbers deployed in warehouses across the USA, although the overall numbers remained small compared with those for stationary and portable fuel cells (U.S. Department of Energy 2014). Fuel cell buses have been commercially available for several years and their usefulness has been well demonstrated. However, their cost, at around five times of a diesel bus, plus the cost of hydrogen infrastructure, mean that they are only used where a city deems the environmental benefit to be worth the extra investment (Ruete 2009). Fuel cell cars are currently only available for lease; these vehicles are being made available by manufactures to gain experience ahead of a commercial launch planned from 2015. In the past decade, PEMFC and DMFC have dominated the total market share in the portable, stationary and transport sectors. (Kendrick 2014) Their uptake by consumers has been facilitated by the development of codes, standards and government policies to lower the barriers to adoption; such as allowing methanol fuel cartridges on board aircraft and feed in tariffs for fuel cell CHP installations.

The use of hydrogen technology may also assist in the introduction of hydrogen obtained from renewable energy sources in the rest of the economy. Andrews and Shabani (2012) have argued in the paper “Re-envisioning the role of hydrogen in a sustainable energy economy” that hydrogen has a critical role to play in a sustainable energy economy based essentially on energy efficiency and renewables, serving as the energy storage needed to guarantee continuous supply, and as transportable fuel to replace petrol and diesel. They also mentioned the new solid-state hydrogen storage technology of metal hydrides is an attractive way to solve the safety problem, and to achieve competitive levels of energy storage per unit mass and volume.
Hydrogen has the highest energy to mass ratio of all fuels (Blencoe 2010). 1 kg of hydrogen contains the same amount of energy as 2.1 kg of natural gas or 2.8 kg of gasoline. On the other hand, hydrogen has a very poor volumetric energy density at normal pressures. The storage of hydrogen is a major problem that has to be solved before the hydrogen economy can begin to be implemented on a large scale. Nowadays, the common ways to store hydrogen are as compressed gas, liquid hydrogen, and in the solid state in metal hydrides. Department of Energy USA (2012) suggests in Fuel cell School Buses Report to Congress, if hydrogen is carried and used on board a vehicle, clearly safety considerations are crucial. On ignition, hydrogen burns in the air as at volumetric hydrogen concentrations from 4 to 74.5 %. Nancy (2012) reports that fuel cell technologies have already been applied in hydrogen station, fuel cell vehicles, distributed generation, forklifts, and backup power.

Ivan (2013) introduce a light electric vehicle for golf carts. This electric vehicle with 5 kW motor was integrated with a commercial 1.2 kW PEM fuel cell system, and the fuel was supplied by two compressed hydrogen cylinders. The introduction of the fuel cell was shown to result in extending the driving range by 63%-110% than the original one without fuel cell, when the amount of stored H2 fuel varied within 55-100% of the maximum capacity. The operation in the hybrid powering mode resulted in more stable driving performance, as well as in the increase of the total energy both withdrawn from and returned to the vehicle battery during the driving (Tolj 2013).

Over the last two year, many companies have released new hydrogen fuel cell cars for commercial sale. Toyota has kept developing their Mirai hydrogen fuel cell car, and it is now on sale in USA and Japan (Toyota 2017). Hyundai has released their ix35 fuel cell powered SUV, with a 100 KW power output, top speed of 160 kph (Hyundai 2017). Mercedes-Benz has developed their F800 fuel cell vehicle. (Mercedes-Benz 2017). Earlier BMW demonstrated a i8-based hydrogen fuel cell research vehicle (BMW 2016).

### 3.1.2 Proton exchange membrane fuel cells
3.1.2.1 Brief history of proton exchange membrane fuel cells

Proton exchange membrane (PEM) technology was invented at General Electric (GE) in the early 1960s, through the work of Thomas Grubb and Leonard Niedrach (Lucia 2012). GE announced an initial success in the mid-1960s when the company developed a small fuel cell for a program with the U.S. Navy's Bureau of Ships (Electronics Division) and the U.S. Army Signal Corps. The unit was fuelled by hydrogen generated by mixing water and lithium hydride. This fuel mixture was contained in disposable canisters that could be easily supplied to personnel in the field. The cell was compact and portable, but its platinum catalysts were expensive.

PEM technology served as part of NASA's Project Gemini in the early days of the U.S. piloted space program (NASA 2010). The lunar flights envisioned for Project Apollo required a power source of longer duration other than the batteries provided in earlier Project Mercury. Gemini's main objective was missions lasting up to 14 days included operational tests of fuel cells. GE's PEM cells were selected, but the model PB2 cell encountered repeated technical difficulties, including internal cell contamination and leakage of oxygen through the membrane. GE redesigned their PEM cell, and the new model P3, despite malfunctions and poor performance on Gemini 5, served adequately for the remaining Gemini flights. GE continued working on PEM cells and in the mid-1970s developed PEM water electrolysis technology for undersea life support, leading to the US Navy Oxygen Generating Plant. The British Royal Navy adopted this technology in the early 1980s for their submarine fleet.

In the late 1980s and early 1990s, Los Alamos National Lab and Texas A&M University experimented with ways to reduce the amount of platinum required for PEM cells (Fuel Cell Today 2013). Overall PEM fuel cells have a mixed record in space over these decades (GE 2015), but several companies also began testing and using these cells in terrestrial vehicles. In 1995, Ballard Systems tested PEM cells in buses in Vancouver and Chicago and later in experimental vehicles made by DaimlerChrysler (Ballard 2013). PEM cells have also supplied power to unmanned blimps called aerostats and to son buoys, which are nautical buoys that generate and receive sonar signals (Fuel Cell Today 2013). Early in 2000, Aero Vironment selected PEM technology to provide night-time power for its solar-powered Helios long-duration aircraft (NASA 2001). The goal was to make an unpiloted aircraft that
could fly continuously for up to six months by using photovoltaic panels during the day to run electric motors and electrolyse water to produce hydrogen gas. At night, the fuel cell was to run the motors by converting the hydrogen and the oxygen back into water. Several test flights were made with and without a fuel cell from 2001 to 2003 (Fuel Cell Today 2013).

Automotive fuel cell research has taken on new urgency as air quality regulations grow steadily stricter, particularly in California. In 2013, Energy Partners and the U. S. Department of Energy's Office of Advanced Automotive Technologies provided two 20 kW fuel cell stacks to Virginia Tech and Texas Tech universities to evaluate their performance in hybrid electric cars. (U. S. Department of Energy 2013) Major automakers like Ford and Volkswagen are also testing PEM vehicles. Recently Toyota, Hyundai and Mercedes Benz have all released new and improved fuel cell vehicles with much higher performance PEM fuel cell stacks (Toyota 2017; Hyundai 2017; Mercedes-Benz 2017; and BMW 2016).

3.1.2.2 Basic structure of PEM fuel cells

The standard single PEM fuel cell is a combination of two endplates as current collectors, two gas diffusion layers, two catalyst layers and a proton exchange membrane.

Hydrogen is fed to the cell through channels cut in the inner surface of one of the end plates. The hydrogen gas passes into the porous gas diffusion layer, which spreads it over the catalyst layer as evenly as possible to form the anode – the negatively polarised electrode – of the cell. An electrochemical reaction happens in this catalyst layer. In which hydrogen
molecules are split on catalyst particles into protons and electrons. The protons pass through the membrane to the cathodic catalyst on its other side. The electrons are not able to pass through the membrane, but instead are conducted through the gas diffusion layer and the external circuit connecting the terminals of the cell to the other endplate. These electrons then travel through the cathodic gas diffusion layer to the catalyst layer on this side of the cell. The oxygen gas is also fed through the pores of the cathodic gas diffusion layer to this second catalyst layer, where it reacts with the protons and the electrons to form water. The movement of the electrons in the external circuit is the current generated by the potential difference that arises between the two end plates. Hence the cell has generated an electrical energy output.

3.1.2.3 Main components and materials

3.1.2.3.1 Membrane electrode assemblies

The heart of a PEM fuel cell is the membrane electrode assembly, which include the membrane, and the catalyst layers on each side. The catalyst layers normally are very thin layer (in the order of $10^{-3}$ mm) and are deposited on both sides of the membrane.

The proton exchange membrane is typically Nafion, a per sulphonated tetrafluoro-ethylene film, which when hydrated serves as a solid polymer electrolyte that conducts only positively charged ions (protons) and blocks the passage of electrons. This polymer electrolyte membrane is the key to PEM fuel cell technology.

Nafion of various chemical compositions and thickness have become the standard material used in PEM fuel cells. But over the last decade, other materials with similar properties have been to overcome disadvantages of Nafion such as its high cost, susceptibility to some gas crossover, maximum temperature of operation of 80 C, and degradation of its structure over time (Grodzinski 2007). For example, nafion has been modified with micron or submicron inorganic/organic additives such as silica (Di Noto et al 2006), TiO$_2$ (Chen et al 2007), TiSiO$_4$ (Devrim et al 2013) and ZrO$_2$ (Jalani, Dunn and Datta 2005). These additives exhibit a high-water retention capability, which allows the membrane to retain proton conductivity at elevated temperatures. Such composite membranes have exhibited improvements in water
retention and PEMFC performance at elevated temperatures. The addition of an inorganic material into polymer membrane can alter and improve physical and chemical polymer properties of interest (such as elastic modulus, solvent permeation rate, tensile strength, hydrophilicity, and glass transition temperature) while retaining its important polymer properties to enable operation in the fuel cell (Peng et al. 2005).

A layer of catalyst is added on both sides of the membrane—the anode layer on the hydrogen side and the cathode layer on the oxygen side. Conventional catalyst layers include nanometer-sized particles of platinum dispersed on a high-surface-area carbon support. This supported platinum catalyst is mixed with a proton-conducting polymer (an ionomer usually nafion,) and is sandwiched between the membrane and the GDLs. On the anode side, the platinum catalyst enables hydrogen molecules to be split into protons and electrons. On the cathode side, the platinum catalyst enables oxygen reduction by facilitating its reaction with the protons generated at the anode that pass through the membrane, and the associated electrons that pass through the external circuit, to produce water. The ionomer mixed into the catalyst layers allows the protons to travel through these layers.

In fuel cells, the electro catalytic reaction, apart from being a function of chemical composition and the electrochemical properties of the catalyst, heavily depends on the catalyst loading itself. A larger catalyst loading enables higher surface area for electrochemical reactions, thus involving more reactant species into the reaction, generating more current and decreasing the activation loss. The catalyst loading is more critical on the cathode side due to the significant activation polarisation/kinetic loss for the oxygen reduction reaction (ORR). Thus, graded catalyst loading, especially at the cathode, controls the electrochemical ORR and the oxygen consumption, resulting in a uniform current density distribution. Furthermore, the high cost of the platinum (Pt) catalyst requires improvements in the catalyst layer performance and catalyst utilisation. Several experimental and numerical studies have been performed to investigate the effect of catalyst layer composition on the performance of fuel cells. The optimum nafion content in the catalyst layer was investigated in the paper of Antolini et al. (1999) and the paper of Xie et al. (2001), while the effects of a cathode catalyst layer with graded Nafion content along the thickness of the layer were studied by Xie et al. (2005) and Wang et al. (2004). The influence of agglomerate size and ionomer distribution on reaction rate distribution and effectiveness of Pt catalyst utilization
was investigated in the paper of Sadeghi, Putz and Eikerling(2014). Qi and Kaufman (2003) experimentally investigated the effect of catalyst loading, Nafion content, cell temperature and electrode drying temperature on the power density of PEM fuel cells. It was found that maximum power density occurred for a Nafion content of 30% and 0.20 ± 0.05 mg/cm² of platinum loading, also the maximum power density was found to increase linearly with increase cell temperature. The effects of design parameters in the catalyst layers such as Nafion content, Pt loading, thickness and porosity of catalyst layers on the cell performance were numerically studied by Song et al (2004) and Srinivasarao et al. (2010). Song et al (2004) used two-parameter optimizations to determine the best combination of two parameters (out of four) to maximize current density, whereas single parameter optimization was studied in the paper of Srinivasaro et al. The effects of catalyst loading on the PEM fuel cell performance were examined in the paper of Smirnova et al. (2005), which showed that cell performance could be maintained with a low catalyst loading could be achieved using a treated novel membrane. Novel carbon aerogel-supported Pt catalysts with varying pore size distributions and Pt content were synthesised and tested in the paper of Smirnova et al. (2005). Antoine et al. (2000) and Ticianelli et al. (1988) studied the effects of catalyst loading along the thickness of the cell. It was found that the optimal catalyst utilization occurs for a thin porous active layer and with Pt nanoparticles close to the proton exchange membrane side. Wilkinson and Pierre (2003) investigated graded catalyst loading along the flow direction, suggesting enhanced performance for high catalyst loading at the inlet. More recently, Cetinbas et al. (2014) numerically investigated the effects of bidirectionally-graded composition, with the catalyst loading variation along the thickness and in-plane direction, for cathode catalyst, and showed that a higher catalyst and/or ionomer loading at the membrane-catalyst layer interface improved the performance.

3.1.2.3.2 Gas Diffusion Layers

The Gas Diffusion layers (GDLs) sit between the catalyst layers and the endplates, aiming at facilitating transport of reactants into the catalyst layer, as well as removing the product water. Each GDL is typically composed of a sheet of carbon paper in which the carbon fibres are partially coated with polytetrafluoroethylene (PTFE) (Park, Lee & Popov 2012). Gases diffuse rapidly through the pores in the GDLs. These pores are kept open by the hydrophobic PTFE, which prevents excessive water build-up. In many cases, the inner surface of the GDL
is coated with a thin layer of high-surface-area carbon mixed with PTFE, called the microporous layer. The microporous layer can help adjust the balance between water retention (needed to maintain membrane conductivity) and water release (needed to keep the pores open so hydrogen and oxygen can diffuse into the electrodes) (Park, Lee & Popov 2012).

However, effective water management is necessary in order to meet fast response to the power required in a system. Deficient water reduces ionic conductivity in the membrane and the catalyst layer as well as induces severe contact resistance between the membrane and the catalyst layer, whereas excessive water in the membrane –electrode-assembly (MEA) reduces catalyst sites for electrochemical reactions and impedes transport of reactants through the non-reactive regions. Liquid water condensed from the water vapor and produced by oxygen reduction reaction at the cathode catalyst layer moves into the membrane or the gas diffusion layer. In the former case, higher liquid water pressure formed by electro-osmotic drag and electrochemical reaction at the interface between the membrane and the catalyst layer drives water flow toward the anode. In the latter, liquid water accumulates at the catalyst layer/GDL interface and the flows toward the gas flow channel when liquid water pressure exceeds a threshold pressure for water flow through the GDL determined by its pore geometry and hydrophobicity.

The GDL in PEM fuel cells in sandwiched between the catalyst layer and the gas flow channel and its structure controls the catalyst utilisation and the overall cell performance. The GDL consists of a macroporous substrate or a thin carbon layer on a sheet of macroporous carbon cloth or carbon paper. It allows gas transport toward the catalyst layer, while allowing liquid water produced at the active catalytic sites to leave the catalyst layer/membrane interface. The GDL must also be a good electrical conductor to allow electrons to flow from the endplate to the catalyst sites on the air/oxygen side of the cell, and from catalyst sites on the hydrogen side. The GDL is typically wet-proofed (usually by PTFE coating) so that the surface and pores in the GDL are not clogged with liquid water that could impede gas transport to the catalyst layer.

3.1.2.3.3 End plates and bipolar plates
The end plates of a single PEM fuel cell serve two roles for the whole cell. First, they are the current collectors in electrical contact with the GDLs on both sides of the cell, allowing electrons produced or consumed in electrochemical reactions to travel from one electrode to the other through an external circuit. Secondly, the end plates also contain the gas channels machined into their inner surfaces. End plates usually are metals, since they should have both good electrical conductivity and physical strength.

As the single PEM fuel cell is operating between 0 to 1.2 V, normally numbers of cells are connected in series to achieve a higher voltage. The connection between adjacent cells is made by a bipolar plate, which in essence is comprised of two end plates (one anodic and other cathodic), set back to back and integrated into a single plate with gas flow channels on both sides. Like endplates bipolar plates thus need to be good electrical conductors, because currents in fuel cell stacks are usually high and even a small ohmic voltage drop across a bipolar plate compared with an average 0.7V operating voltage results in significant energy losses. The anode side and cathode side of the next layer fuel cell are constructed in one plate, which has gas channels on both sides. Bipolar plates are made from a variety of materials including graphite, stainless steel, and coated aluminium. Modern PEM fuel cells are increasingly using plates made of pressed metal and then welded together on their flat surfaces to reduce the total mass of the stack.

3.1.2.4 PEM fuel cell stacks

PEM fuel cell stacks are formed by stacking a series of single fuel cells with each cell connected to the previous one by bipolar plates, and end plates at the beginning and end. The voltage of a stack is the sum of the voltages of the individual cells, while the same current flows through each cell in the stack. Hence the power output rises linearly with the number of cells connected in series, as long as the output per cell remains the same.

Ballard Company provides several kinds of fuel cell stacks, classified as air-cooled and liquid-cooled. The air-cooled fuel cell stacks are 400 W to 3.3 kW, with 43W for each cell. The liquid-cooled fuel cell stacks are 3.8 kW to 21 kW. This cell stack is air-cooled and larger fuel cell stacks is liquid-cool. Ballard also has Fuel Cell Modules in 30 kW to 200W. for forklifts, buses and light rail applications. Nedstack provides 2.0-10kW HP fuel cell stack,
designed for non-continuous operation at high currents up to 230A, or XXL type for continuous operation. Horizon provides a huge range of different power output from 50W to 5kW.

3.1.2.5 New developments in PEM fuel cells

Silica particle is a typical example of inorganic filler that according to their size, shape, distribution, amount and orientation within nano- or micro-composite membranes applied to PEM fuel cells, can meet various technological requirements for electrolyte fuel cells, such as enhanced proton conductivity, reduced methanol permeability and improved mechanical stability. In Devrim’s paper (2015), he designed an air-cooled PEM fuel cell short stack with nafion/silica nanocomposite membrane and fabricated for net 100W net power output to improve the stack performance at low relative humidity conditions. Devrim believes humidifier not only contributes to the system’s weight and cost, but is also a parasitic load, resulting in a reduced efficiency. To overcome the hydration limitation of the nafion membrane, modifications of these membranes with micron or submicron inorganic/organic additives such as silica, TiO₂, TiSiO₄ and ZrO₂ have been investigated. These additives exhibit a high water retention capability, and can enhance the membrane’s capacity to retain high hydration and hence high proton conductivity at elevated temperatures. Devrim obtains such a new idea, and provides a lot of parameters of each part of the fuel cell. Nafion membranes could be modified to achieve significant for the application at low humidity or elevated temperatures. To overcome the hydration limitation of the nafion membrane, alternative polymeric membranes, such as nanocomposite membranes, have been investigated (Fu et al 2008). Gokce, Berker and Inci (2016) studied the influence of fluorinated ethylene propylene nanoparticles in catalyst layer on water management and performance of PEM fuel cell with high Pt loading. Antonio et al (2016) present their work on optimization of a PEM fuel cell operating conditions: Obtaining the maximum performance polarization curve. Their work was done on a 50cm² PEM fuel cell. Ali et al (2016) had studied two-phase flow and droplet behaviour in microchannels of PEM fuel cell. Afshari, Dehkordi and Rajabian (2016) present an investigation of the PEM fuel cells performance with partially restricted cathode flow channels and metal foam as a flow distributor.
3.2 COMPUTER SIMULATION MODELS AVAILABLE

Fritts and Savinell (1989) developed a mathematical model to describe the performance of hydrogen electrode catalyst particles bonded to a proton exchange membrane of a hydrogen-bromine fuel cell. This paper presented an early simulation of a fuel cell in a 1D mathematical model with values of key parameters (like cell current) aggregated across each plane. The model used the Butler-Volmer equation to represent the interfacial reaction rates on each electrode. This 1-D model cannot show any variations in parameter across planes parallel to the active area of the membrane. In addition, these early models did not include the liquid water formation, which is very important in limiting the maximum current generated by the cell.

The difficulty in performing experiments to determine variation in reaction rates across the active membrane area and the actual dynamics of liquid water formation in a PEM fuel cell have motivated researchers to develop computational simulation models to obtain qualitative insight, and ultimately quantitative analysis, of these key factors affecting fuel cell performance. Early two-phase flow modelling approaches applied to PEM fuel cells, representing water in both vapour and liquid phases, were the multi-phase mixture (M2) model and the multi-fluid model (also called two-fluid model, Golpaygan and Ashgriz 2008). The M2 model includes liquid water saturation as a volume fraction of liquid water, and the multi-phase mixture is considered to be a single-phase fluid with a varying phase composition. The M2 approach was first used to model the two-phase flow in PEM fuel cells by Wang et al. (2001). Other examples of two-phase PEM fuel cells models using this approach include those developed by You and Liu (2002), Mazumder and Cole (2003) and Pasaogullari and Wang (2005). Multi-fluid models were developed by He et al. (2007), Ye and Van Nguyen (2007), Berning (2008) and Gurau et al. (2008). Le et al. (2010) assumed that the mixture in the M2 model did not have separate liquid and gas phases, and that the multiphase flow was obtained through a mass-averaged mixture velocity.

In contrast to the M2 model, Anderson et al. (2010) developed a multi-fluid model in which liquid and gas phases were represented by one complete set of conservation equations (mass, momentum and energy), and the two phases were coupled by the saturation state. Ferreira et
al. (2014) concluded in their review that since the multi-fluid approach had only a few a priori assumptions it was more realistic than the earlier models. Moreover, the phase transfer equilibrium between the liquid and gas phases was also taken into account to describe condensation and evaporation. However, the multi-fluid model required a high number of dependent variables, and can be challenging to couple the phases to achieve stable solutions.

Ferreira et al. (2014) stated in their review paper that both the M$^2$ and multi-fluid schemes are able to consider both liquid and gas phase transport and can thus predict the amount of liquid water (as liquid water saturation) present in a PEM fuel cell and its effects on cell performance. However, these models cannot distinguish between the presence of liquid water as liquid droplets or water films locally concentrated, dispersed, or accumulated in the gas channels. Therefore, interface tracking algorithms are needed for the formation and motion of liquid water cannot be modelled. Several numerical simulation methods for tracking multiphase flows across interfaces have been applied to PEM fuel cell research, namely the Lattice Boltzmann (LB), the pore-network (PN), the level set (LS) and the volume of fluid (VOF) methods (Ferreira 2014). The LB method, unlike the conventional Navier-Stokes solvers based on the macroscopic (tens of microns to meters) continuum equations, considers the fluid to be composed of a collection of pseudo-particles, which perform consecutive propagation and collision processes over a discrete lattice mesh (Ferreira 2014). Such particulate kinetics provides a relatively easy and consistent way to consider microscopic interactions. In the PN model, a porous medium is represented by a lattice of wide pores connected by narrower constrictions called throats. This modelling approach can be less computationally intensive while accounting for all the relevant two-phase flow physics (Ferreira 2014). Unfortunately, as mesoscopic models (less than microns), the LB and PN schemes are only applicable at the micro-scale, and they are not suitable for simulating the macro-scale two-phase flow patterns that occur in the gas channels of a PEM fuel cell, where variations take place typically at the tens of micron scale (Ding et al 2011). Thus, studies employing these approaches have been focused mostly on water-gas micro-scale transport in the porous media of the cell.

The macroscopic scale from tens of microns to meters is, however, well suited for modelling by the LS and VOF continuum methods and computations based on conservation laws. (Worner 2012) The LS method is relatively simple to implement and is capable of handling
topological changes and complex interfacial shapes in a simplified way. A serious issue with this model is its susceptibility to mass conservation errors which requires sophisticated measures to be avoided. Nevertheless, some researchers have employed the LS approach to study the two-phase flow in PEM fuel cells (Choi et al. 2009, Akhtar and Kerkhof 2011). The VOF method with interface reconstruction, on the other hand, is strictly mass conservative. This technique is able to simulate immiscible fluids by solving a single set of momentum equations and then tracks the volume fraction of each fluid throughout the domain. Moreover, it is capable of considering surface tension and wall adhesion effects. In addition, the VOF method is incorporated as a ready-to-use model in many commercial computational fluid dynamics (CFD) software packages, including ANSYS Fluent, GERRIS Flow solver (GFS), and STAR-CCM. These features make the VOF method the most popular model to simulate the two-phase flow phenomena in PEM fuel cells.

In table 3.1, some key papers over the past ten years on fuel cell modelling, have been selected and classified according to the dimensionality of the geometric representation in the model, the approach to representing fluid phases, whether variation of parameters by finite element over space or dealing just with aggregated parameters, and computational software employed.

<table>
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<th>Year</th>
<th>Author</th>
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<td>3D</td>
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Notes:

(1): Multiphase here means liquid (water), plus gas (reactants, and the reaction product, water vapour)
(2): Parameters like current, gas flows, heat production aggregated over the full active area of the membrane
(3): Fluent, Inc. was acquired by ANSYS, Inc. in 2006, and is now part of the ANSYS software package.

Table 3.1: Selected studies of PEM fuel cell modelling over the past ten years

Maher et al. (2007) presented a comprehensive three-dimensional multi-phase computational fluid dynamics model of a PEM fuel cell that allowed temperature variations over space and represented the significant physical processes and the key parameter affecting fuel cell performance the using a three-dimensional, multi-phase, non-isothermal model. The model accounted for both the gas and liquid phase in the same computational domain, and thus could represent phase change inside the gas diffusion layers. The model included the transport of gaseous species, liquid water, protons, energy and water dissolved in the ion conducting polymer membrane. Water was assumed to be exchanged among three phases; liquid, vapor and dissolved in the membrane, and equilibrium among these phases is assumed. The model featured an algorithm that provided a more realistic representation of the local activation over-potentials, which led to improved prediction of the local current density distribution. This model also took into account convection and diffusion of different species in the channels as well as in the porous gas diffusion layer, heat transfer in the solids as well
as in the gases and electrochemical reactions. The influence of various parameters on fuel cell performance, including geometry, materials, operating and others, was modelled. In addition, the model was capable of identifying critical parameters for the wetting behaviour of the gas diffusion layers and could be used to determine conditions that might lead to the onset of pore plugging, which has a detrimental effect on the fuel cell performance.

In Maher et al.’s (2007) paper, a full three-dimensional, multi-phase computational fluid dynamics model of a PEM fuel cell with straight flow channels had been developed. The model was developed to improve fundamental understanding of transport phenomena in PEM fuel cells and to investigate the impact of various operation parameters on performance. The major transport phenomena in a PEM fuel cell accounted for: namely convective and diffusive heat and mass transfer, electrode kinetics, transport and phase change mechanism of water, and electric (potential) fields. The model accounted for the liquid water flux inside the gas diffusion layers by viscous and capillary forces, and hence, was capable of predicting the amount of liquid water inside the gas diffusion layers. The physics of phase change were included by prescribing the local evaporation term as a function of the amount of liquid water present and the level of under saturation, whereas the condensation was a function of the level of over saturation. The new feature of the algorithm developed in this work was its capability for accurate calculation of the local activation overpotentials, which, in turn, resulted in the improved prediction of the local current density distribution.

Maher et al. (2007) applied this comprehensive model to an optimisation study that sought to quantify the impacts of operating, design and material parameters on fuel cell performance. The model was able to: (1) improve understanding of the many interacting, complex electrochemical and transport phenomena that cannot be studied experimentally; (2) identify limiting steps and components; and (3) provide a computer-aided tool for design and optimisation of future fuel cells potentially with much higher power density and lower cost. In addition, the model was capable of identifying important parameters for the wetting behaviour of the gas diffusion layers and could be used to determine conditions that might lead to the onset of pore plugging in GDLs, which has such a detrimental effect on fuel cell performance, especially in the mass transport limited region.

Maher et al.’s (2007) paper described the development of the model, the determination of properties for use in the model, the validation of the model using experimental data and the application of the model to explain observed experimental phenomena. A computational
model of an entire cell requires vast computing resources and excessively long simulation times. The computational domain in this study was therefore limited geometrically to one straight flow channel and adjacent electrical contact areas on each side (cathode and anode) of the membrane electrode assembly. The multi-phase model was validated by comparing its results to experimental data in a published paper (Maher et al. 2007). The values of the electrochemical transport parameters for the materials employed and the base-case operating conditions were listed. It is important to note that because this model accounted for all major transport processes and the modelling domain comprised all the elements of a complete cell, no parameters needed to be adjusted in order to obtain physical results. The comparison of the cell potential versus current density curve obtained from the experimental results with that from the multi-phase 3D model showed close agreement.

The governing equations were discretised using a finite volume method and solved using a general purpose computational fluid dynamic code. Stringent numerical tests were performed to ensure that the solutions were independent of grid size. A computational mesh of 150,000 computational cells was found to provide sufficient spatial resolution. The detailed solution procedure is described in Maher et al. (2007). The strength of their model was that it clearly allowed parametric studies to be performed, and hence the impacts of various parameters on the transport mechanisms and overall fuel cell performance to be explored. The feature of the algorithm developed in this work was its capability for accurate calculation of the local activation overpotentials, which, in turn, resulted in improved prediction of the local current density distribution.

Lin et al. (2007) developed a quasi-two-dimensional mathematical model for a polymer electrolyte membrane fuel cell with consideration of axial convection in the gas channel and analytical solutions were obtained. A half-cell model which contains the cathode gas channel, gas diffuser, catalyst layer, and the membrane is investigated. To account for the effect of gas velocity in the gas channel, axial convection is included in the oxygen transport equation of the gas channel. Expressions for the oxygen mass fraction distribution in the gas channel, gas diffuser, and catalyst layer, and the current density and the membrane phase potential in the catalyst layer and membrane are derived. The solutions are presented in the form of convergent infinite series. The polarisation curve is also expressed as a function of the surface overpotential. Due to the advantage of the closed-form solutions this model can be
easily employed as a diagnostic tool for PEM fuel cell simulations. It was found that for a given oxygen concentration on the channel wall the variation of oxygen mass fraction along y axis with the effect of axial convection is more uniform and larger in the gas channel than that obtained in one-dimensional studies, which showed a linear profile across the channel. The larger oxygen mass fraction leads to larger mass fraction in the GDL and catalyst layer and the resulting current density is higher. The overall performance at low overpotential was controlled by the surface kinetics in the catalyst layer. As the overpotential increased, the electrochemical reactions became more active, the oxygen mass fraction decreases exponentially along the channel, and the induced current density was larger. For higher overpotential, electrochemical reactions occurred only in the very thin layer situated at the interface between the catalyst layer and the gas diffuser and the corresponding current density reached its maximum value in this very thin layer and then remains constant further inside. When the overpotential was over 0.35V, the distribution of oxygen mass fraction remained unchanged with overpotential in the whole domain, and the corresponding current density and performance did not increase further. The limiting situation was reached and the overall performance was controlled by the rate of mass diffusion/convection. The oxygen mass fraction had higher values in the downstream region for larger values of the inlet gas velocity. The resulting profiles of mass fraction therefore decay more gradually in the axial direction. In the region downstream, mass fraction increased with velocity and so did the loss of phase potential, the corresponding potential loss and current density are larger, and the performance of the fuel cell improves but more oxygen is exhausted from the cell. The value of the limiting current density was independent of the properties related to the catalyst layer, and increased with the inlet gas velocity. In this paper, the axial convection was considered only in the gas channel, and the gas velocity was assumed constant; diffusion in the Y direction was the only mode of transport in the gas diffuser layer and the catalyst layer; the system considered is steady and isothermal; the air is full saturated with water vapor and taken as an ideal gas; the membrane is maintained in a fully hydrated condition; oxygen is transported to the catalyst sites as a gaseous component only, and liquid water does not constitute a continuous barrier to oxygen transport. So this model is a simplified model to avoid too many variable parameters during the operation. As a result, the accuracy of the model may be limited, but it still could be used to study the effects of various input parameters while keeping other parameters constant.
Baschuk and Li (2009) presented a comprehensive, consistent and systematic mathematical model for PEM fuel cells that can be used as the general formulation for the simulation and analysis of PEM fuel cells. As an illustration, the model is applied in this paper to an isothermal, steady state, two-dimensional PEM fuel cell. Water is assumed to be in either the gas phase or as a liquid phase in the pores of the polymer electrolyte. The model includes the transport of gas in the gas flow channels, electrode backing and catalyst layers; the transport of water and hydronium in the polymer electrolyte of the catalyst and polymer electrolyte layers; and the transport of electrical current in the solid phase. Water and ion transport in the polymer electrolyte was modelled using the generalized Stefan–Maxwell equations, based on non-equilibrium thermodynamics. Model simulations showed that the bulk, convective gas velocity facilitated hydrogen transport from the gas flow channels to the anode catalyst layers, but inhibited oxygen transport. While some of the water required by the anode was supplied by the water produced in the cathode, the majority of water must be supplied by the anode gas phase, making operation with fully humidified reactants necessary. The length of the gas flow channel had a significant effect on the current production of the PEM fuel cell, with a longer channel length having a lower performance relative to a shorter channel length. This lower performance was caused by a greater variation in water content of the longer channel length.

The governing equations were solved using the finite volume method with computer codes developed in-house. Model simulations illustrated three important phenomena in PEM fuel cells. The gas phase flow in the gas flow channels, electrode backing and catalyst layers showed that there was a net mass transport from the anode side of the cell to the cathode side. This net mass transport created a bulk velocity that favours hydrogen transport from the gas flow channels to the anode catalyst layers, but inhibited the flow of oxygen to the cathode reaction sites. Water was consumed in the anode catalyst layer and produced in the cathode catalyst layer. Some of the water required by the anode was transported from the cathode to the anode through the polymer electrolyte. However, the gas phase of the anode side supplied the majority of the water, making operation with fully humidified reactants necessary. The length of the gas flow channel had a significant effect on the current production of the PEM fuel cell, with a longer channel length having a lower performance relative to a shorter channel length. This lower performance was caused by a large water content variation in the fuel cell with the longer channel length. The operating parameters could be changed during the typical operation of a PEM fuel cell. The operating parameters correspond to the
boundary conditions of the governing equations and the cell operating temperature. The computational domain included the physical layers of the fuel cell, such as the gas flow channels, electrode backing catalyst and polymer electrolyte layers. Also included in the computational domain are extensions of the gas flow channels, referred to as the ‘plus’ and ‘minus’ extensions. These extensions were not physically part of the PEM fuel cell, but are included to facilitate the numerical solution of the governing equations. As this is pure mathematical model, there is no comparison between experiment data. However, this model was used to show effects of some variables, which would be used to guide the further study work.

Martins et al. (2009) introduced a simplified and comprehensive PEMFC mathematical model in order to predict the response of PEM fuel cells to the variations in physical properties of manufactured materials, and in operating and design parameters. The model consists of the conservation equations for each control volume, and the equations accounting for electrochemical reactions. The reversible electrical potential and power of the fuel cell are then computed as functions of the temperature and pressure fields determined by the model. This PEM fuel cell simulation model with temperature dependence on space and current was then experimentally validated by direct comparison with output voltage and power measurements performance in PEMFC’s in the laboratory. The model accounts for pressure drops in the gas channels, and temperature gradients with respect to space in the flow direction. Current density variations are investigated experimentally by direct infrared imaging, showing that even at low current operation such gradients are present in fuel cell, and therefore should be considered by a PEMFC model, since large coolant flow rates are limited due to induced high pressure drops in the cooling channels. The computed polarisation and power curves are directly compared to the experimentally measured ones with good qualitative and quantitative agreement. In Martins’ opinion, it is desirable that the model for the fuel cell structure optimisation should be as simple as possible, while still capturing the most important physical phenomena present in the fuel cell, to keep the computational time low to provide solutions for each tested configuration. This experimental rig had 10 single PEM fuel cells manufactured with the same specifications to provide the internal and external geometry for simulation, as well as actual physical properties of the manufacturing materials, including density, porosity, specific heat and electrical conductivity. There were some necessary data taken form the technical literature. Since the values for the
anode and cathode exchange current densities are commonly reported over a wide range, the values adopted in his study were determined by the solution of an inverse problem of parameter estimation, by entering each measured voltage and current in the mathematical model, and calculating exchange current density as unknowns. The average of all values calculated in that manner were within the accepted range for PEMFC anode and cathode exchange current densities reported by the literature. Overall Martins et al. model was found to be able to simulate some aspects of the performance in the PEM fuel cell, although it was concluded that more work would be needed to improve the detail and accuracy of the modelled cell performance.

Both steady-state and dynamic behaviour of a PEM fuel cell is modelled and simulated by Sharifi Asl, Rowshanzamir and Eikani (2010). Two mathematical models were developed one for computing the steady-state, and another for dynamic voltage–current (V–I) characteristics of PEM fuel cell stacks. For determining the humidity of the membrane in steady-state conditions, theoretical equations were derived and used. The goal of dynamic modelling is to find the response of the system to variations in the load. In this research, in addition to the charge double-layer phenomenon, the effects of temperature and gas flows were taken into account. The fuel cell system is divided into three control volumes and thus a lumped-parameter model for these sub-systems is established using the mass and heat transfer equations. The proposed models are implemented in a Matlab/Simulink environment. Using the dynamic form of the model, the transient response of voltage under sudden change in load current was studied. Pure hydrogen and air are supplied to the anode and cathode channel, respectively. In addition to output voltage profiles, the dynamic changes over time of anode and cathode pressure, hydrogen mass in anode channel, oxygen and nitrogen mass in the cathode channel, inlet hydrogen, oxygen and nitrogen flow rate, and stack temperature were predicted, and compared with experimental data on commercial fuel cells and simulation results of other researchers.

Srinophakum and Markumchan (2011) used molecular dynamics simulation for ionic conductivity in a chitosan membrane for a PEM fuel cell. A classical molecular dynamics simulation using a COMPASS force field was applied to systems containing chitosan, hydronium ions, and various amounts of water: 10, 20, 30 or 40% water. The simulation
predicted the diffusion coefficient, the ion conductivity, the coordination between particles. The system containing 40% water was found to be most suitable as a conducting material because it had the highest ionic conductivity $\left(7.14 \times 10^{-2} \, \text{S/cm}\right)$. Such material was studied in a temperature range of 298–360 K, and the conductivity results obtained followed Arrhenius’ behaviour. A study at 70, 80, 90° of deacetylation (DDA) at a constant of 40% water content showed the same trend in conductivity behaviour. The coordination was considered in evaluating the location of particles in order to understand the transport mechanism of the ions. Conductivity is appropriate in the systems in which the Eigen ion water cluster was formed. This work presented the ion conductivity mechanism of a chitosan membrane based on a molecular level. Firstly, the structures of each particle were formulated and minimized to obtain a stable conformation.

In the paper of Falca et al. (2011), one-dimensional and three-dimensional numerical simulations were performed and compared with experimental data obtained in a PEM fuel cell. The 1D model, coupling heat and mass transfer effects, was previously developed and validated by the same author. There were some assumptions used to avoid too many variables. The 3D numerical simulation was conducted using the commercial code FLUENT –PEMFC module, which is also used in the present project. The geometry of the fuel cell included the MEA and the channel plates. Terminal plates and collector plates were not considered, in order to achieve shorter simulation times. The depth of channel in each side was different. In this model, the electrochemical processes were treated as heterogeneous reactions taking place on the catalyst surfaces. Two electrical potential fields are solved: one in the membrane and catalytic layers and the other in the catalytic layers, diffusion layers and current collectors. Surface reactions occurring on the porous catalyst region are solved and the reaction diffusion balance is applied to compute the rate. The current density value is computed based on a cell voltage prescribed. To shorten computing time, the Tafel equation was used in all the simulations, as opposed to the full. Butler-Volmer equations that will be used in this thesis (chapter 4). Falca et al.’s, results showed that 1D and 3D model simulations considering just one phase for the water flow were similar, with a slightly better accordance for the 1D model exhibiting a substantially lower CPU time. However, both numerical results for 1D model overpredicted the fuel cell performance while the 3D simulations reproduced very well the experimental data. The effect of the relative humidity of gases and operation temperature on fuel cell performance was also studied both through the
comparison of the polarization curves for the 1D and 3D simulations and experimental data and through the analysis of relevant physical parameters such as the water membrane content and the proton conductivity. A polarisation curve with the 1D model was obtained with a CPU time around 5 min, while the 3D computing time is around 24 h. The results showed that the 1D model can be used to predict optimal operating conditions in PEMFCs and the general trends of the impact on fuel cell performance of several important physical parameters (such as those related to the water management). The use of the 3D numerical simulations were needed for more detailed predictions including the spatial distribution of key parameters like current density, gas concentration and water saturation spatial visualisation of various relevant parameters. An important conclusion of this work is the demonstration that a simpler model using low CPU has potential to be used in real-time PEMFC simulations. 3D simulation work takes a much longer time, but is necessary to develop a detailed spatially-disaggregated analysis of key parameters in a PEM fuel cell and a PEM fuel cell stack. Importantly, the ANSYS Fuel Cell Module used in the present project is basically the same as this Fluent PEMFC module reported by Falca et al. (2011).

In Meidanshahi and Karimi’s paper (2012), a nonlinear one-dimensional along-the-channel dynamic model is investigated for modelling and simulation of power generation in a PEM fuel cell. The proposed model is based on conservation laws (energy, mass and species), and electrochemical and auxiliary equations. The operating parameters of the cell are optimised under the steady state condition to obtain the best system performance. Based on the proposed mathematical model, a fuzzy-logic controller is designed to control the average power density in the fuel cell. The fuzzy controller is applied to the process and the results are compared with those of a tuned conventional Proportional-Integral (PI) controller. Differential evolution is used as the optimization algorithm and the inlet molar flow rates of hydrogen and oxygen, inlet temperatures of anode and cathode gases, inlet molar flow rate of coolant, and inlet pressures of anode and cathode gases are optimised as the operating parameters. The fluxes in y-direction are taken into account as they affect the concentrations of reactants and products in the gas channel, but the changes of concentrations of reactants and products in y-direction are not considered with the exception of water flux through the membrane. This model is mathematical model simulates the performance of whole cell dealing with aggregate values such as total current. Although this model could help to
achieve better fuel cell performance, it is not able to represent spatial variation in the plane of the membrane.

In the study of Hashemi, Rowshanzamir and Rezakazemi (2012), a comprehensive non-isothermal three-dimensional model has been established to investigate the performance of proton exchange membrane fuel cells with both straight and serpentine flow fields. The model considers the major transport phenomena in a fuel cell involving mass, heat and energy transport, electrode kinetics, and potential fields. Oxygen and hydrogen mass fraction distributions, current density and temperature distribution are determined. The activation overpotentials anode and cathode are constant. Modelling findings showed that profiles differ from those simulations that assume constant activation overpotential. The model equations were solved by a numerical method based on finite volume. The model found that the serpentine flow field showed more even distributions of current density and temperature across the active area of the cell. Simulation results were compared with the experimental data reported in literature and global comparisons showed good agreement between the model and experimental results. The simulation model described in this paper is very similar to the ANSYS Fuel Cell Module that is used in the present thesis.

Ding, Bi and Wilkinson (2013) used a 3D volume fluid model coupled with a 1D membrane electrode assembly model to investigate the impact of two-phase flow patterns, especially slug flow, on fuel cell performance. The proposed model only dealt with two-phase flow on the cathode side. A 1D MEA model is adopted to simplify the transport across the GDL, the catalyst layer and the membrane, and the two-phase flow in the 3D gas flow channel is simulated by the VOF method. Hence only the gas flow is represented in 3D, not other processes within the cell like current flow. The MEA model and the 3D flow channel model are coupled at GDL surface with lies between the gas flow channel and the MEA. Several pores at the GDL surface are used to represent the large pores inside the GDL where liquid water emerges. The model described in this paper is not a complete fuel cell model, simulating only a single cathode channel coupled with the GDL and MEA, as their focus is water management. The water flux calculated in the 1D MEA model contains both liquid water and water vapor. Typically, the water evaporation rate in the gas channel was fast enough to saturated the gas if liquid water was present there. Therefore, it is assumed that the
water generated by the reaction first humidifies the gas to saturation. This mount of water directly enters the channel at its local boundary, and if there is still water left, it will emerge from the nearest pore in the GDL as liquid. On the other hand, if water vapour somewhere in the channel is oversaturated, the excess water vapour will condense, and be added into the nearest emerging pores as well. This model is validated against experimental results by comparing the polarization curves obtained from experiments and simulations. The two-phase flow hydrodynamics in the 3D gas flow channel was simulated by the VOF method, and the commercial software FLUENT was used in the simulation.

Bilgili, Bosomoiu and Tsotridis (2014) modelled gas flow fields with obstacles in PEM fuel cells under different operating conditions. In PEMFCs laminar flow inside anode and cathode gas channels can be disrupted by using obstacles. By arranging these obstacles near to the exit of the gas channels concentration losses due to hydrogen and oxygen consumption inside the channel will be decreased. Bilgili, Bosomoiu and Tsotridis (2014) use a three-dimensional computational model to perform numerical simulations of the performance of PEMFCs containing obstacles in the anode and cathode gas flow channels. The conservation equations in all layers of the cell were solved using a finite-volume computational fluid dynamics code in Fluent. The CFD code included a module for PEM fuel cells adapted to calculate the source terms for heat, species transport, and electrochemical reactions. This model is almost the same as that used in the present work for a fuel cells with a single pair of channels in chapter 4.

In Headley and Chen’s paper (2015), the necessity of multiple thermal control volumes (CVs) in the cathode channel is investigated to address the spatial gradients in temperature. A method to improve the thermal modelling accuracy near the cathode inlet of the fuel cell stack by using un-evenly spaced CVs to represent the channel is presented. Analysis is performed starting with the 1D heat transfer equation for the cathode channel. By using scaling arguments to remove extraneous terms, a simplified 1D solution for the temperature profile in the cathode channel is developed. From this result, the necessary CV sizes to remain within a critical band of the actual spatial temperature profile is calculated. The result of this study gives a good basis for a similar analysis of the relative humidity profile in the fuel cell stack. To perform a reasonable scaling analysis of the humidity evolution, thorough
understanding of the temperature evolution is necessary. Future work planned by these researchers will include an impact analysis of the quick temperature change on the performance of the inlet cell. Also, a similar analysis will be conducted to characterise and size control volumes for accurate relative humidity modelling of the cathode channel, to gain improved understanding of localised dehydration and flooding effects. The fast, dynamic thermal model was developed to simulate the temperature response in the anode, cathode, and coolant channels, and fuel cell body. Experimental inlet data were used as the inputs to the model. This data included the inlet flow rates, pressures and temperature for each channel, and the current demand. The main outputs calculated by the model are the outlet temperatures and stack voltage. This model is a thermal dynamic model, with electric current as one of the input parameters, in contrast the model used in the project where spatial distribution of current density, and hence total cell current, are outputs of the model.

Temperature distribution in a single PEM fuel cell is studied by Cao et al. (2015) using a 3D non-isothermal numerical model with gas diffusion layers with anisotropic properties. The model is applied to investigate the effect of thermal contact resistance (TCR) between rib and GDL, channel and rib width, and different heat exchange coefficients, on the temperature distribution and performance of the cell. The thermal contact resistance in this model is determined by comparing the predicted temperature variation between plate and cathode electrode to the available experimental results. The numerical results proved that to improve the prediction accuracy of temperature distribution and cell performance, the effect of TCR cannot be neglected. An underestimate of temperature of 1.5 K is found when the cell output voltage was 0.6 V, compared to the case without TCR. And it is found that the rib and channel width and the ratio between them have an obvious effect on heat and mass transfer processes occurring in the electrode. A near optimal rib-to-channel width ratio was found to be 1.0 mm/0.8 mm, and the narrower the channel and rib widths the better the performance when their widths are equal. By comparing the temperature distributions obtained with different assumed heat exchange coefficients for gas diffusion layers, it was found that, when natural air convection is applied to cooling down the PEM fuel cell, the generated heat cannot be removed completely. However, when liquid water is used as the cooling fluid, the heat removal capacity greatly exceeded the real demands of a single PEM fuel cell, so that the water must be heated to prevent over cooling of the fuel cell. The computational domain in this study is a typical unit ‘part-cell’ within a parallel flow plate, comprising half of a gas
channel and collector rib. This basic unit is then repeated periodically. The model used in the present project will, however, cover the full active area without breaking this up into assumed equal unit ‘part cells’.

Macedo et al (2016) have developed a three-dimensional model to simulate the fluid flow, heat transfer, electrochemical reaction and species transport in a Proton Exchange Membrane Fuel Cell stack with five single cells including the membrane, gas diffusion layers, catalyst layers, flow channels and current collectors. A commercial computational fluid dynamics (CFD) code, Ansys 15.0®, was used to solve the numerical model. The numerical results showed the detailed distributions of mass fractions of hydrogen and oxygen, as well as the heat sources and temperature through five membrane electrode assemblies inside the stack. The species concentration always was higher at inlets and it was reduced gradually along the channels. The lowest temperature was observed at the inlet of the cathode where oxygen is supplied at temperature of 300 K. Likewise, the heat sources in PEM fuel cell are tightly related with the current density distributions through membrane electrodes assembly. Moreover, the results were validated by polarisation curves and experimental data obtained from experiments carried out in a Horizon PEM fuel Cell (H- 100). The PEM fuel cell stack model was developed using CFD commercial code. Numerical simulations were conducted for a stack with five single cells connected in series. The numerical results showed that the model could predict the temperature variation, as well the temperature distribution through the membrane electrode assembly and through each cell inside the stack. In this case, the higher temperature values are shown at ribs of the cathode and near to the outlet flow of the cathode, and the lower temperature values are shown at the inlet. Contours of the reaction heat source and the ohmic heat source were also presented. This model is an important step for further development of the stack design and the flow field of a PEM fuel cell stack, and is very similar to that used in the present thesis and reported in chapter 6. But the configurations of the gas flow channels used in this thesis are different to those investigated by Macedo et al. (2016), and most importantly the gas supply for each layer of cell is independent in the modelling conducted here.

As this review has shown, mathematical models of PEM fuel cell have been developed and used since the late 1980s. Since this time, the models have been developed very fast with a trend towards increasing sophistication and comprehensiveness in terms of physical,
chemical, electrochemical and electrostatic processes covered quantitatively. Most models have sought to represent the fluid dynamics of the PEM fuel cell, including hydrogen and air flows, and formation of water. Since the late 2000s finite element analysis, including Computational Fluid Dynamics, has been employed to model input and output gas flows. The electrochemical processes, relationships between electric potential, generated EMFs, and current flows, and heat generation and transfer, were in the early years dealt with on an aggregated basis over the full active area of the membrane, rather than in terms of spatially varying parameters across this active area. A particularly challenging task has been the accurate modelling of two-phase flows, which is representing the actual dynamics of liquid water in a PEM fuel cell. Early two-phase flow modelling is considered to be a single-phase fluid with a varying phase composition. These models did not have separate liquid and gas phases, and the multiphase flow is obtained with a mass-averaged mixture velocity. It was not until 2007 that a fuel cell model was developed that took into account the phase transfer equilibrium between the liquid and gas phase through condensation and evaporation. Even though the models have become increasingly more realistic, they still cannot identify the water as liquid droplets or water films locally.

Yet since around 2010, more sophisticated models have been developed to deal with all of the following processes, and the links between them, in a truly 3D manner in single PEM fuel cells:

- the fluid dynamics in the cell, incorporating the actual geometries of flow channels and representations of porous media, and usually employing a CFD simulation package
- the dynamic balance between liquid water formation, water vapour concentration (that is, related to relative humidity and cell temperature), and air flows both into and out of the cell
- the relationships between electric potential and current densities within all the various media in the cell (end plates, gas diffusion layers, catalyst layers and the membrane as functions of 3D position
- the generation of heat from the electrochemical reactions and heat transfer.

In recent years, some models of PEM fuel cell stacks, covering some or all of these processes, have also begun to appear. But to date, fuel cell stack modelling has invariably involved
some simplifying assumptions, such as independent gas supply to each cell layer, or equal potential differences across each cell in stack. Hence future model development will no doubt concentrate on relaxing these constraints and obtaining variations in conditions and performance cell within the stack as a crucial output of the modelling process.

3.3 SELECTION OF ANSYS FUEL CELL MODULE FOR THE PRESENT STUDY

This project seeks to investigate the influence of active membrane area and stacking PEM fuel cells in a stack on overall performance. To take account of as many potentially relevant influences as possible, the modelling needs to:

- Be three dimensional, representing the actual geometry of the fuel cell, including the gas flow channels, the area and thickness of the membrane, and the properties of the catalyst and gas diffusion layer.
- Simulate multi-phase flow, including water formation in the catalyst layer, and water saturation in gas diffusion layers and catalysts.
- Take account fluid dynamics as well as spatial variation of the electrical potential, current density, gas flow rates, gas pressures and relative humidity.
- Calculate temperatures in the cell and the associated heat transfers.
- Be based on finite elements to show the spatial distribution of all key parameter such as current density, temperature, gas concentration and pressure, and water saturation.
- Be applicable to fuel cell stacks, show any differences in performance from cell to cell within the stack.

The review in this chapter has shown that a number of potentially suitable models have already been developed over a period of ten years or more, with some experimental testing of these. Hence it was decided not to develop a completely new model in the present project.

Consideration of the available models, shows that those based on Matlab/Simulink and SIMPLE focus on a specific topic of interest, and cannot meet the requirement of this project.
to represent an entire cell or stack, layer by layer, in a spatially-disaggregated manner. For example, Sharifi et al.’s (2010) Matlab/Simulink model focusses on the voltage-current characteristic of the cell and its dependence on average temperature and gas flow. Hashemi et al.’s (2012) model based on SIMPLE made simplifying assumptions such as that all generated water exists in the gas phase, and that the membrane is completely hydrated with a fixed proton conductivity.

The ANSYS Fluent general PEM fuel cell module (ANSYS 2012) has therefore been selected for use in the present project for the following main reasons:

- ANSYS Fluent accepts 3D spatial models, so that the exact geometry of our experimental fuel cells, in terms of thickness of catalysts, gas diffusion layers, membrane, and configuration of gas flow channels, can be fully represented.

- The ANSYS Fluent Fuel Cell Module can model the liquid water formation and transport in the porous media of the cell, including the catalyst and gas diffusion layers.

- This Module computes the electric field throughout the cell layers and uses this field to calculate current density, and hence ohmic heating in all conducting solid and porous regions of PEM fuel cell.

- The Module shows the distributions through space of current density, gas concentrations, gas flows, and temperature throughout the PEM fuel cell.

- The Module is also able to represent fuel cell stacks with realistic 3D geometry, and variations in gas supply and hence current density distribution from cell to cell.

3.4 PREVIOUS USE OF ANSYS FUEL CELL MODULE TO STUDY DIFFERENT DESIGN OPTIONS FOR SINGLE PEM FUEL CELL AND FOR STACK MODELLING
The ANSYS Fuel Cell Module was introduced in Chapter 3.3, and has already been used for several years, mainly for flow-field design and water management. There have been many recent papers using ANSYS Fluent to model liquid water in flow channels, the heat exchanger for PEM fuel cells, and high-temperature PEM fuel cells. In this section, only studies specifically using the ANSYS Fuel Cell Module are reviewed.

Guo et al. (2013) used the ANSYS Fuel Cell Module to study the flow field of a bipolar plate focussing on reactant gas and removal of the produced water. A pin-type flow field configuration was modelled, which has the advantage of a low pressure drop at the expense of uneven flow distribution and stagnant areas. Guo et al. (2013) developed a network-based optimisation model was to optimise this flow-field configuration. Two cases uniform flow distribution with and without considering reactant consumption along flow channels, were investigated and the corresponding optimised designs developed. Three-dimensional numerical simulations were conducted to confirm the proposed optimisation model and also to compare the performance of the fuel cells using the optimised designs and the regular pin-type configuration. Both the optimised designs exhibited substantially higher fuel cell performance compared to the regular pin-type configuration. Moreover, the design with an optimised flow field obtained by considering reactant consumption performed better than the one without considering this consumption. The current density distribution in the membrane was used to evaluate the performance of different design of the flow channel.

Han et al. (2015) reported on an ANSYS Fuel Cell Module study of the performance and flow characteristic of a large PEM fuel cell having channels like tree branches. Increasing the active area of each cell allows reduction in the number of cells in a stack to obtain a given level of power output, and possible gains in cost effectiveness. This study investigated flow channel configurations that could be sued in cells with a large active area. Computational analysis showed that, in the case where a branching factor (f) of the branch channel is changed from 1 to 0.5 in its exit part, the performance of the channel was similar, but the pressure drop was reduced by 78% compared to a serpentine channel. Experimental tests suggested that, while the serpentine channel produced 139.8 W of electrical power, of the associated blower consumed 9.12 W of electric power to overcome the pressure drop. But in the case of the branch channel with the f value of 0.5 in its exit part, the blower consumed
only 4.38 W, which was a 3.55% greater performance compared to the serpentine channel. The comparison between simulation and experiment was not enough to validate the simulation result.

An ANSYS Fuel Cell Module study by Saco et al. (2016) compared various flow channel configurations and effective water management to optimise power output. To compare these configurations and water management strategies, experimental work would have been considerably more time consuming and costly. Saco et al. (2016) were able to use the ANSYS Fuel Cell Module to do this comparison for a PEMFC with an active area of 225 cm². It was found that the power density developed by straight flow channel with zig-zag flow path is 0.3758 W/cm² and is the maximum of the configurations considered due to effective water management with minimal pressure drop.

Gomez et al (2014) used the ANSYS Fuel Cell Module with some added time-dependent functions to model the effect of operating parameters on the transient performance of polymer electrolyte membrane fuel cell stack with a dead-end anode. The operation of polymer electrolyte membrane fuel cell (PEMFC) stack with a dead-end anode requires careful consideration on the gas and water management. Water accumulation at the anode and the nitrogen crossover from cathode to anode led to performance deterioration over time. The accumulated water and nitrogen was removed by purging to ensure good and stable stack performance. This study aimed at the experimental and numerical evaluation of the effect of the key operating parameters – inlet humidification, stoichiometry, and operating current – on the transient performance of a dead-ended anode fuel cell stack. The experiments were carried out on a stack with 24 cells and a catalyst active area of 300 cm². By employing a validated transient two-phase mathematical model of a PEMFC with a dead-end anode, numerical simulations were performed that yielded a better understanding of the local distribution of water, and hydrogen, oxygen, water vapor and nitrogen. The results suggested that the deterioration in fuel cell performance over time is closely related to the choice of the operating conditions. The anode and cathode inlet conditions became a limiting factor for stack performance. Liquid accumulation at the anode was found to be strongly related to the inlet air humidification as well as water transport across the membrane, whereas the cathode stoichiometry affected the nitrogen crossover. By contrast, in the present project, only steady-
state conditions will be modelled using the ANSYS FC Module, for which it is primarily designed.

### 3.5 GAPS IN KNOWLEDGE AND UNDERSTANDING

From the foregoing review a number of gaps in knowledge and understanding relating to the simulation modelling of PEM fuel cells and stacks emerge as follows:

- There is a lack of full 3D modelling taking into account gas flows, two-phase flow regions, electrochemical reactions, and charge flow (electrons and protons) in all regions of single PEM fuel cells, with detailed comparison of the simulation results with experiments conducted on a cell of the same design and materials. Maher’s (2007) model had one channel on each side and no endplate. Maher compared his model with other’s experimental data. The input setting of electrode and membrane parameters were from different reference. Falca’s model also did not consider the endplates. The model compared with the experimental data in voltage versus current density curve. Hashemi’s model was also quite simple, and liquid phase was on considered. Macedo’s fuel cell stack model was focus on heat distribution and gas concentration, without a comparison with experiment.

- Few papers have focussed on the effects of operating conditions – reactant flow rates, cell and gas temperatures, gas pressures, relative humidity, mass fraction of liquid water – on cell performance. Baschnick studied the transport of gas and water. Martin’s model focus on the temperature. Meidansha interested in the gas flow. Ding’s model was on water only.

- Little work has been done to compare the performance (per unit membrane active area) of large and small single cells, with a detailed analysis of the variations in current density across the active area and the causes of these variations.

- There is a lack of understanding of and empirical data on the causes of the turndown in power output at high current densities, relating to saturation of porous GDLs, gas channels, and catalyst layers with liquid water, and kinetic limitations in the electrochemical reactions.
There has so far been relatively little experience gained in the detailed 3D modelling of PEM fuel cell stacks in which the inflows to and outflows from, and voltage of, each cell can vary. Maced’s model was a 5-layer fuel cell stack.

The present thesis will seek to make a contribution to improving knowledge and understanding in all these areas.
4. THE ANSYS FUEL CELL MODULE:

SETTING UP FOR INVESTIGATION OF

A PEM FUEL CELL

4.1 ANSYS FUEL CELL MODULE

4.1.1 Introduction to ANSYS Fuel Cell Module structure and software

In this project, the ANSYS Fuel Cell Module has been used to represent the actual geometry of and physical and electrochemical conditions in single PEM fuel cells and stacks. Hence the module is applied to model every part of the fuel cell, and stacks of individual cells, in three dimensions. The computation fluid dynamics tool - Fluent - from ANSYS can simulate fluid flows at both the cell and stack level, and by using numerical modelling can be used to optimise the fluid mechanical aspects of fuel cell design to achieve superior power generation rates, durability and reliability. Fluent also can help in assessing the role of thermal and electromechanical stresses on cell and stack performances.

Specifically the ANSYS Fluent Fuel Cell Module contains component models of the following process in a PEM fuel cell:

- The electrochemical reactions that take place in the catalyst layers of a PEM fuel cell are simulated by the electrochemistry models to speculate local current density and voltage distributions. Electrochemical models consider the losses, including activation loss, ohmic loss and concentration loss.

- Liquid water flows in the fuel cell are predicted by a multiphase mixture model. Water is produced because of the reaction between protons (hydronium H₃O⁺), oxygen fuel and electrons from the external circuit at the interface between the MEA
and the cathode gas diffusion layer. Water is also transported through the membrane due to electro-osmotic drag. If the cathode gas flow is saturated, this water is produced in liquid form. As liquid water reaches the surface of the diffusion layer, it is introduced into the gas flow passage as a thin liquid film of drops, where shear between the gas flow and film surface can move the film. The re-entry of liquid water from the thin film back into the diffusion layer is also permitted. The multiphase mixture model from ANSYS solves for conservation of mass, momentum, energy and species in the porous diffusion layer. ANSYS tools utilises local values of the liquid volume fraction – that is, the fraction of any infinitesimal volume that is occupied by liquid water – within porous media to describe liquid water flows. The resulting effect on local void fraction is used to modify the local value of gas phase diffusivity, thermal conductivity and electrical conductivity throughout the cathode diffusion layer. This modification to the transport properties for local water concentration accurately models the effect of water production on mass diffusion, heat conduction and electrical conduction (ANSYS 2014).

- The net transfer of liquid water and electrical losses across the membrane-electrode assembly – consisting of the thin polymer membrane and catalyst layers – are calculated. Liquid water travels across the membrane due to electro-osmotic drag and molecular diffusion. The simulation tools calculate the protonic conductivity of the MEA as a function of both water concentration and temperature.

- The electric potential fields are calculated by ANSYS and then used to predict local current density, voltage and ohmic heating in all conducting solid and porous regions of PEM fuel cells.

- Contact resistances at appropriate interfaces can also be set.

The heart of the electrochemistry models is the computation of the rates of the anodic and cathodic reactions, using Butler-Volmer type equations. In these equations, the driving force behind the reactions is the surface over-potential: the difference between the phase potential of the solid catalyst and the phase potential of the electrolyte/membrane. Two potential equations are solved in the ANSYS module: one potential equation accounts for the electron transport through the solid conductive materials, and the other potential equation represents the protonic transport for H⁺. The following equations in this subsection (equations 4.1 to 4.19) are all taken from ANSYS FLUENT Fuel Cell Module Manual (2012).
\[ \nabla \cdot (\sigma_{sol} \nabla \phi_{sol}) + R_{sol} = 0 \]  \hspace{1cm} (4.1)

for electron flow through the solid material of catalyst layer and GDL, and

\[ \nabla \cdot (\sigma_{mem} \nabla \phi_{mem}) + R_{mem} = 0 \]  \hspace{1cm} (4.2)

for proton flow through the membrane,

where, \( \sigma \) = electrical conductivity (1/ohm-m);

\( \phi \) = electric potential (volts);

\( R \) = volumetric transfer current (A/m\(^3\)).

The transfer currents, or the source terms in the above equations, are non-zero only inside the catalyst layers and are computed as:

for the solid phase, \( R_{sol} = -R_{an}(< 0) \) on the anode side and \( R_{sol} = +R_{cat}(> 0) \) on the cathode side; and

for the membrane phase, \( R_{mem} = +R_{an}(> 0) \) on the anode side and \( R_{mem} = -R_{cat}(< 0) \) on the cathode side.

The current density source terms, \( R_{mem} \) and \( R_{sol} \) (A/m\(^3\)), have the following general definitions:

\[ R_{an} = \left( \xi_{an} J_{an}^{ref} \right) \left( \frac{[A]}{[A]_{ref}} \right)^{\gamma_{an}} \left( -e^{+\alpha_{an} F \eta_{an}/RT} + e^{-\alpha_{cat} F \eta_{an}/RT} \right) \]  \hspace{1cm} (4.3)

\[ R_{cat} = \left( \xi_{cat} J_{cat}^{ref} \right) \left( \frac{[C]}{[C]_{ref}} \right)^{\gamma_{cat}} \left( -e^{+\alpha_{an} F \eta_{cat}/RT} + e^{-\alpha_{cat} F \eta_{cat}/RT} \right) \]  \hspace{1cm} (4.4)

where, \( J_{an/cat}^{ref} \) = reference exchange current density per active surface area (A/m\(^2\))

\( \xi \) = specific active surface area (1/m)

\( [A],[C]_{ref} \) = local species concentration, reference value (kmol/m\(^3\))

\( \gamma \) = concentration dependence exponent

\( \alpha \) = exchange coefficient (dimensionless)

\( F \) = Faraday constant (9.65 \times 10\(^7\) C/kmol).
Species volumetric source terms (kg/m$^3$-s) in the catalyst layer, also called the triple-phase boundaries (TPBs), due to electrochemical reactions for the PEMFC are:

$$S_{H_2} = -\frac{M_{w,H_2}}{2F} R_{an} < 0 \quad (4.5)$$

$$S_{O_2} = -\frac{M_{w,O_2}}{4F} R_{cat} < 0 \quad (4.6)$$

$$S_{H_2O} = \frac{M_{w,H_2O}}{2F} R_{cat} > 0 \quad (4.7)$$

Additional volumetric sources in the thermal energy equation are present because not all chemical energy released in the electrochemical reactions can be converted to electrical work due to the irreversibility of the process. The net energy available is given by the equation:

$$S_h = h_{react} - R_{an,cat} \eta_{an,cat} + I^2 R_{ohm} + h_L \quad (4.8)$$

where $h_{react}$ is the net enthalpy change due to the electrochemical reactions, $R_{an,cat} \eta_{an,cat}$ is the product of the transfer current and the overpotential in the anode or the cathode TPB, $R_{ohm}$ is the ohmic resistivity of the conducting media, $h_L$ is the enthalpy change due to condensation/vaporisation of water, and $I$ is the total cell current.

Since PEM fuel cells operate at a relatively low temperature of around 50°C, the water vapour may condense to liquid water, especially at high current densities. While the existence of the liquid water keeps the membrane hydrated, it also blocks the gas diffusion passages, reduces the diffusion rates and the effective reacting surface areas and hence limits the cell performance. To model the formation and transport of liquid water, ANSYS Fluent use a water saturation model (ANSYS 2012).

The liquid water formation and transport is governed by the following conservation equation for the volume fraction of liquid water, $s$, or the water saturation.

$$\frac{\partial (\epsilon \rho s)}{\partial t} + \nabla \cdot (\rho \nabla s) = r_w \quad (4.9)$$

where,

$$r_w = c_r \max \left( \left[ (1 - s) \frac{P_{w,v} - P_{sat}}{R_T M_{w,H_2O}} \right], [-s \rho_l] \right) \quad (4.10)$$

where $-r_w$ is added to the water vapor equation, as well as the pressure correction (mass source). This term is applied only inside the catalyst and gas diffusion layers. The
condensation rate constant was hardwired to \( c_r = 100 \text{s}^{-1} \) according to the manual of ANSYS Fuel Cell Module, which was not able to be changed. It is assumed that the liquid velocity, \( V_l \), is equivalent to the gas velocity inside the gas channel. Inside the highly-resistant zones, the use of the capillary diffusion term allows us to replace the convective term in equation 4.9:

\[
\frac{\partial (\varepsilon \rho_l s)}{\partial t} + \nabla \cdot \left[ \rho_l K s^3 \frac{dp_c}{ds} \nabla s \right] = r_w
\]  (4.11)

Depending on the wetting phase, the capillary pressure is computed as fraction of \( s \),

\[
p_c = \begin{cases} 
\frac{\delta \cos \theta_c}{\varepsilon} \left( 1.417 (1-s) - 2.12 (1-s)^2 + 1.263 (1-s)^3 \right), & \theta_c < 90^\circ \\
\frac{\delta \cos \theta_c}{\varepsilon^{0.5}} \left( 1.417 s - 2.12 s^2 + 1.263 s^3 \right), & \theta_c > 90^\circ 
\end{cases}
\]  (4.12)

where \( \varepsilon \) is the porosity, \( \delta \) is the surface tension (N/m\(^2\)), \( \theta_c \) is the contact angle and \( K \) the absolute permeability.

Gas phase species diffusivities are computed by using the dilute approximation method as:

\[
D_i = \varepsilon^{1.5} (1-s) r_s D_i^0 \left( \frac{p_0}{p} \right)^{\gamma_p} \left( \frac{T}{T_0} \right)^{\gamma_t}
\]  (4.13)

where \( D_i^0 \) is the mass diffusivity of species of \( i \) at reference temperature and pressure \((p_0, T_0)\). These reference values and the exponents \((\gamma_p, \gamma_t)\) as well as the exponent of pore blockage \( (r_s) \) are defined in the Fuel Cell and Electrolysis user defined functions (UDF) as,

\[
p_0 = 101325 \text{ N/m}^2
\]

\[
T_0 = 300 \text{ K}
\]

\[
\gamma_p = 1.0
\]

\[
\gamma_t = 1.5
\]

\[
r_s = 2.5
\]

The electrolyte phase conductivity, also called membrane conductivity, is modelled as:

\[
\sigma_{mem} = \beta (0.514 \lambda - 0.326) \omega e^{1268 \left( \frac{1}{303.17} \right)^{m}}
\]  (4.14)
where $\lambda$ is water content. Two model constants, $\beta$ and $\omega$ are introduced in ANSYS Fluent for generality.

The osmotic drag coefficient is modelled as:

$$n_d = 2.5 \frac{\lambda}{22} \quad (4.15)$$

The back diffusion flux of water is modelled as:

$$J_{w \text{ diff}} = -\frac{\rho_m}{M_m} M_{H_2O} D_l \nabla \lambda \quad (4.16)$$

where $\rho_m$ and $M_m$ are the density and the equivalent weight of the dry membrane.

The membrane water diffusivity is modelled as:

$$D_l = f(\lambda)e^{2416\left(\frac{1}{303} - \frac{1}{T}\right)} \quad (4.17)$$

The water content of the membrane is modelled as:

$$\lambda = 0.043 + 17.18\alpha - 39.85\alpha^2 + 36\alpha^3 (\alpha < 1) \quad (4.18)$$

$$\lambda = 14 + 1.4(\alpha - 1)(\alpha > 1) \quad (4.19)$$

where $\alpha$ is water activity defined as: $\alpha = \frac{P_{wv}}{P_{sat}} + 2s$.

The water vapour pressure is computed based upon the vapour molar fraction and the local pressure,

$$P_{wv} = x_{H_2O} P \quad (4.20)$$

The specific leakage current, $I_{\text{leak}}$ (A/m3), models the effect of species crossover from one electrode to another across the electrolyte.

$$S_{H_2} = -\frac{M_{w,H_2}}{2F} I_{\text{leak}} \quad (4.21)$$

$$S_{O_2} = -\frac{M_{w,O_2}}{4F} I_{\text{leak}} \quad (4.22)$$

$$S_{H_2O} = \frac{M_{w,H_2O}}{2F} I_{\text{leak}} \quad (4.23)$$

Neither the general nor detailed procedures used in the ANSYS Fuel Cell Module to perform its calculations are available to users. It is likely, however, that the basic flowchart of the calculation method is similar to that shown in Figure 4.1.
4.1.2 Inputs required

The first step in an ANSYS Fuel Cell Module project is to define the geometry of the various regions of the cell. A three-dimensional drawing can be made in ANSYS Geometry. This program is most useful for a simple structure but not so easily applied for a complex structure. However, the required three-dimensional drawing can also be imported from other commercially-available software, such as Solidworks or AutoCAD, which is more suitable for complex structures. An example of using ANSYS Geometry to define and input the geometry of a basic fuel cell with a single pair of gas flow channels is described in later in section 4.3. The geometries of the small-scale single-layer multi-channel fuel cell, and other larger fuel cells, investigated in this project were drawn in Solidworks and subsequently transferred to ANSYS.
The second step is to set up mesh to be employed in the various regions of the fuel cell, that is, dividing up each region into small repeating ‘unit shapes’ that fill the entire space of the cell. The specific boundaries between regions must also be defined and named in the meshing procedure. Meshing is a complex task requiring considerable experience, as the dimensions of the mesh should be small enough to reflect differences in key parameters to a sufficient degree of precision along all three axes, and at the same time large enough to keep the computing time acceptably short. For example, for the small single fuel cell (active membrane area of 5 cm$^2$) investigated in this project using a refined high-quality (that is, small-volume) mesh, the computing time for each voltage point with the same input parameters was typically over 8 hours.

After transferring the meshing data to Fluent, the ANSYS Fuel Cell Module is active as add on-module in FLUENT. There are eight options for model simulation:

- The Joule Heating option takes into account ohmic heating;
- The Reaction Heating option takes into account the heat generated by electrochemical reactions;
- The Electrochemistry sources option allows the Fuel Cell Module to take electrochemistry effect in to account.
- The Butler-Volmer Rate option is used to compute the transfer currents inside the catalyst layers. If this option is turned off, the Tafel approximation is used.
- The Membrane Water Transport option takes into account the transport of water across the membrane.
- The Multiphase option takes in to account multiphase calculations. This option is used for solving approximate liquid transport in the gas diffusion layer of the fuel cell.
- The Multicomponent Diffusion option is used to compute the gas species mass diffusivity using full multicomponent diffusion method.
- The Anisothropic E-Conductivity in Porous Electrode option is used to model the typically non-isotropic electrical conductivity.

The required options must be selected.

Secondly, the input parameters specifically for the Fuel Cell Module are required. For electrochemistry, these are the reference exchange current density, reference concentration, concentration exponent and exchange coefficient is required for both anode and cathode.
(equations 4.3 and 4.4). As well, open-circuit voltage, specific leakage current and reference diffusivity of species must be entered in the appropriate dialog box.

The next step is specifying anode properties by using the anode table of the Module to specify zones and properties of the current collector the flow channel, the diffusion layer, and the catalyst later for the anode portion of the fuel cell. The first properties to be entered are for the current collector. A corresponding zone from the Zone(s) list is selected when Current Collector is selected under the Anode Zone Type. Then a solid material form the corresponding drop-down list is selected. Initially, the material is aluminium, which is not the one used in experiments in this project. A new material should be customised using the materials dialog box. Secondly, for flow channel, the only step is to select a corresponding zone. Thirdly, the selection is named as Porous Electrode, which is also called as gas diffusion layer in this thesis. For this part, besides solid material, porosity, viscous resistance and contact angle are also required. Contact angle is requested only for the multiphase fuel cell calculation.

Fourthly, for the Triple Phase Boundary (TPB) layer (catalyst), besides solid material, porosity, viscous resistance, surface/volume ratio and contact angle are required. Contact angle is requested only for the multiphase fuel cell calculation. Surface/volume ratio of catalysts is used in Butler-Volmer equations. In the electrolyte tab, corresponding zone should be selected as ‘membrane’. Here some properties such as equivalent weight, protonic conduction coefficient and protonic conduction exponent are required. For the cathode side, the setting is similar to the anode side.

For advanced setup, contact resistivity, coolant channel and stack management can be specified independently. Stack management is used to model a fuel cell stack, not for the single fuel cell model in Chapter 4 and 5.

In the tab of ‘reports’, electrolyte projected area is required for calculating the average current density. The External Contact interface(s) fields are the faces of the endplates that act as external contact surfaces for the anode and the cathode.
The following boundary conditions need to be defined for the Fuel Cell and Electrolysis simulation based on problem specification:

- In the ANSYS fuel cell module, the user-defined scalar allocations are termed “uds-\(x\)”. UDS-0 refers to Electric Potential; UDS-1 refers to Protonic Potential; UDS-2 refers to Water Saturation; UDS-3 refers to Water Content.
- Anode Inlet: Mass flow rate; temperature; direction specification method; mass fractions; the coolant must be set to zero if coolant channels are enabled; UDS-2 water saturation must be set to 0.
- Cathode Inlet: Mass flow rate; temperature; direction specification method; mass fractions; the coolant must be set to zero if coolant channels are enabled; UDS-2 water saturation must be set to 0.
- Coolant Inlet (if any) mass flow rate; temperature; direction specification method; coolant mass fraction set to 1; UDS-2 water saturation must be set to 0;
- Pressure Outlets(all): realistic backflow conditions;
- Terminal Anode: Temperature; UDS-0 electric potential set to ground voltage
- Terminal Cathode: temperature; UDS-0 electric potential is set to the voltage of the cathode. If solving for constant current, the UDS-0 flux is set to the current density in A/m2.

For potentiostatic boundary conditions, after initialisation, steady state solutions are calculated for cell voltages close to the open-circuit voltage. The same can be said for galvanostatic boundary conditions and low electric current. By lowering the cell voltage or by raising the average electric current, subsequent stationary solutions can be calculated.

In summary, all the major input parameters are listed Table 4.1.

<table>
<thead>
<tr>
<th>Anode</th>
<th>Unit</th>
<th>Cathode</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ref. Current Density</td>
<td>A/m²</td>
<td>Ref. Current Density</td>
<td>A/m²</td>
</tr>
<tr>
<td>Ref. Concentration</td>
<td>kmol/m³</td>
<td>Ref. Concentration</td>
<td>kmol/m³</td>
</tr>
<tr>
<td>Concentration exponent</td>
<td>Concentration exponent</td>
<td>Concentration exponent</td>
<td></td>
</tr>
<tr>
<td>Exchange Coefficient(a)</td>
<td>Exchange Coefficient(a)</td>
<td>Exchange Coefficient(a)</td>
<td></td>
</tr>
<tr>
<td>Exchange Coefficient(c)</td>
<td>Exchange Coefficient(c)</td>
<td>Exchange Coefficient(c)</td>
<td></td>
</tr>
</tbody>
</table>
### Table 4.1: Input parameters required by the ANSYS Fuel Cell Module

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Overall</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Equivalent Weight</td>
<td>kg/kmol</td>
<td>Open-Circuit Voltage</td>
<td>V</td>
</tr>
<tr>
<td>Protonic Conduction Coefficient</td>
<td></td>
<td>Specific Leakage Current</td>
<td>A/m³</td>
</tr>
<tr>
<td>Protonic Conduction Exponent</td>
<td></td>
<td>Electrolyte Project Area</td>
<td>m²</td>
</tr>
</tbody>
</table>

Other than the settings for these parameters, the properties of the material of each part are also very important and must be input to the model. These material properties are listed in Table 4.2.
Table 4.2: Material properties of cell components required as inputs to the ANSYS Fuel Cell Module

<table>
<thead>
<tr>
<th>Property</th>
<th>Unit</th>
<th>Membrane</th>
<th>Endplate</th>
<th>Catalyst</th>
<th>Gas diffusion layer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness</td>
<td></td>
<td>c</td>
<td>a</td>
<td>c</td>
<td>a</td>
</tr>
<tr>
<td>Density</td>
<td>kg/m³</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cp (specific heat)</td>
<td>j/kg-k</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Thermal Conductivity</td>
<td>w/m-k</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Electrical Conductivity</td>
<td>1/ohm-m</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Basis weight</td>
<td>g/m²</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>Protonic Conduction Coefficient</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>Protonic Conduction Exponent</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>Porosity</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Viscous Resistance</td>
<td>1/m²</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td></td>
</tr>
<tr>
<td>Surface/Volume ratio</td>
<td>1/m</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
<td></td>
</tr>
</tbody>
</table>

The Ref. Current Density, Ref. Concentration, Concentration exponent, Exchange Coefficient, Surface/Volume Ratio are used in the Butler-Volmer equation. Protonic Conduction Coefficient and Protonic Conduction Exponent are used in equation 4.10 to calculate the membrane conductivity.

There are two exchange coefficients on each side. For the early version of Fuel Cell Module, there is only one exchange coefficient on each side. This problem is discussed in the follow section 4.2.
4.1.3 Possible outputs

4.1.3.1 Report as graphics

ANSYS Fluent gives multiple kinds of graphical displays of simulation outputs. In the present project, the most useful graphics are contours graphics and vector graphics.

Contours graphics provide a coloured contour map of the values of a parameter across any planes specified. For each fuel cell model, the planes are defined at the middle of channels, the contact surface of catalysts and gas diffusion layers, and the middle of membrane (across its thickness) to present the simulation result. Contour maps across planes in channels can be used to show the mass fractions of gases, gas flow velocity, pressure and density. The planes between catalyst and gas diffusion layers can be used to show the molar concentration of H₂ and O₂. The planes within the membrane are used to show the current density and water content, such as current density in membrane in Figure 4.2. The surfaces of the two endplates are also used to show the current density.

Figure 4.2: Contour map of current density distribution in membrane
Vectors graphics provide vectors of parameters in any plane or volume specified in the fuel cell. Normally the graphics used in plane to avoid mass arrow of vector in volume. In the following chapters, the vectors of gas flow velocity are presented in the planes of each channel (Figure 4.3), and vectors of current flux density in cross section of the fuel cell through all the layers (Figure 4.4).

Figure 4.3: the graph of velocity of oxygen

Figure 4.4: vectors of current flux density in cross section of the fuel cell through all the layers
4.1.3.2 Reports as numerical results

Important numerical outputs of a simulation are:

- The total cell current, calculated as the integral of the current density vector across either of the endplate surfaces.
- The total mass flow at each inlet and outlet, calculated as a surface integral of flow rate per unit area across the corresponding inlet and outlet surface.
- The average values of mass fraction of both hydrogen and oxygen at each inlet and outlet, calculated as a surface integral of the gas concentration across these surfaces.

The average values of mass flow rate and mass fraction are used to calculate the consumption of each gas. The consumption of gas is used in calculating the energy efficiency of the cell:

- The total transfer current on each electrode, calculated as a volume integral over the catalyst layer.
- The maximum and minimum value of current density across a specified plane, calculated as an integral of current density vectors over all directions.

The integral value of transfer current is used to compare with integral value of current density at the end surface, and hence total cell current. The values of total cell current and current density at a given cell voltage can be compared with experimental value to check the validity of the simulation.

4.1.3.3 Voltage-current characteristic curves and power output

The voltage-current (VI) and voltage-current density (average) (V-J) curves can be plotted by successive runs of the model to find the total current for a given input cell voltage. These characteristic curves provide a convenient comparison with the experimentally-measured performance of a cell.

Estimating the total electrical power output of the cell (the product of cell voltage and current) at various operating points is another important aim of the simulation. This power output together with the corresponding input mass flow rate of hydrogen can be used to calculate the energy efficiency of the cell under varying operating conditions.
4.2 CORRECTION TO BUTLER-VOLMER EQUATIONS IN ANSYS FUEL CELL MODULE

4.2.1 Correction to Butler-Volmer equations

On checking the description in the ANSYS Fluent Fuel Cell Modules Manual (2012) of the use of Butler-Volmer equations to derive the current generation in the oxygen and hydrogen-side catalyst layers in a PEM cell, a possible error in this formulation has been detected. In these equations, it would appear that the same values for the exchange coefficients are used in both the oxygen-side and hydrogen-side equations. But in the usual Butler-Volmer equations, the exchange coefficients used respectively on the two electrodes of a cell are different.

In the ANSYS Fuel Cell Module (ANSYS 2012), the basic Butler-Volmer equations are expressed as follows:

\[
R_{an} = \left( \zeta_{an} j_{an} \right) \left[ \frac{|A|}{|A|_{ref}} \right] \gamma_{an} \left( -e^{+\alpha_{an} F \eta_{an} / RT} + e^{-\alpha_{cat} F \eta_{an} / RT} \right) \tag{4.20}
\]

\[
R_{cat} = \left( \zeta_{cat} j_{cat} \right) \left[ \frac{[C]}{[C]_{ref}} \right] \gamma_{cat} \left( -e^{+\alpha_{an} F \eta_{cat} / RT} + e^{-\alpha_{cat} F \eta_{cat} / RT} \right) \tag{4.21}
\]

where

- \( R_{an}, R_{cat} \): electrode current density \( A/m^2 \);
- \( j_{an}, j_{cat} \): reference exchange current density per active surface area \( A/m^2 \);
- \( \zeta_{an}, \zeta_{cat} \): specific active surface area \( 1/m \);
- \( [A, C], [A, C]_{ref} \): local species concentration, reference value \( kmol/m^3 \);
- \( \gamma_{an}, \gamma_{cat} \): concentration dependence;
- \( \alpha_{an}, \alpha_{cat} \): exchange coefficient (dimensionless);
- \( F \): Faraday constant \( 9.65 \times 10^7 \) C/kmol;
- \( \eta \): activation overpotential.
In these equations, the same values for the exchange coefficients (\(\alpha_{\text{cat}}\) and \(\alpha_{\text{an}}\)) are used on both the oxygen-side and hydrogen-side equations. But in the usual Butler-Volmer equations, the exchange coefficients used respectively on the two electrodes of a cell are different.

This practice is consistent with the definition of the exchange coefficient (for the forward reaction on a particular electrode) as the fraction of the overpotential that is effective in encouraging the forward reaction on that electrode; and similarly for definition of the exchange coefficient (reverse reaction) on that electrode. The values of these coefficients on a given electrode will depend on the catalyst material, structural features of catalyst layer, and the particular reaction that is taking place. Hence there is no a priori reason why the exchange coefficients on one electrode (for forward and reverse reactions) should be equal to the corresponding values on the other electrode.

The Butler-Volmer equations used in the Module should therefore be:

\[
R_{\text{an}} = \left(\zeta_{\text{an}} \frac{[\text{A}]}{[\text{A}]_{\text{ref}}}\right)^{Y_{\text{an}}} \left(-e^{+\alpha_{\text{an}}F\eta_{\text{an}}/RT} + e^{-\alpha_{\text{an}}F\eta_{\text{an}}/RT}\right)
\] (4.22)

\[
R_{\text{cat}} = \left(\zeta_{\text{cat}} \frac{[\text{C}]}{[\text{C}]_{\text{ref}}}\right)^{Y_{\text{cat}}} \left(-e^{+\alpha_{\text{cat}}F\eta_{\text{cat}}/RT} + e^{-\alpha_{\text{cat}}F\eta_{\text{cat}}/RT}\right)
\] (4.23)

The change exchange coefficient \(\alpha_{\text{an, cat}}^{re}\) for the reverse reaction on each electrode can be replaced by \(1 - \alpha\), \(\alpha\) is charge exchange coefficient for the forward reaction define for both \(\alpha_{\text{an}}\) and \(\alpha_{\text{cat}}\).

We have let ANSYS know of this problem and they have accepted this argument and modified the source code in later versions of the module accordingly to correct the problem.
4.2.2 Correction to the software script of the module

Butler-Volmer equations in this modified form of equations 4.22 and 4.23 have thus been incorporated into the ANSYS Module by modifying the source code, and the Module used in this corrected form throughout this thesis.

The source code used to describe the equations for the Fuel Cell Module is in the file pem_user.c, located in “ANSYS Inc\v145\fluent\fluent14.5.0\addons\fuelcells\src\”. The Butler-Volmer equations are used to calculate the transfer current. The code related to Butler-Volmer equations is shown as below:

```
real Transfer_Current(real i_ref, real gamma, int species_i, real alpha_a,
                      real alpha_c, real *dRade, real *dRcde, Thread *t, cell_t c)
```

Purpose: compute transfer current, A/m3, (Ra and Rc in the fuel cell manual)

Input: i_ref - effective transfer current coefficient, computed by
       Cathode_J_TransCoef(c,t) or Anode_J_TransCoef(c,t)
       gamma - cathode or anode concentration exponent
       species_i - species index used in fuelcells (e.g. i_o2, i_h2, i_h2o)
       alpha_a - product of anode exchange coefficient and F/(RT)
       alpha_c - product of cathode exchange coefficient and F/(RT)
       *t - current thread
       c - current cell

Output: source - anode or cathode volumetric transfer current (Ra or Rc)
       *dRade - partial derivative of Ra with respect to activation loss
       *dRcde - partial derivative of Rc with respect to activation loss
...
if (zt == cca_zone)
{
    eta = local_V_open;
    if (FC_Model==PEMFC || FC_Model==SOFC)
        eta = MAX(-local_V_open,MIN(-eta_cut,eta));
/* prevent over- and underflow of exp() */
    alpha_eta_c = alpha_c*eta;
    if (exponent_limit < alpha_eta_c)
        exp_c = exp_big;
    else if (-exponent_limit > alpha_eta_c)
        exp_c = exp_sml;
    else
        exp_c = exp(alpha_eta_c);

if (Butler_Volmer)
{
/* prevent over- and underflow of exp() */
    alpha_eta_a = alpha_a*eta;
    if (exponent_limit < alpha_eta_a)
        exp_a = exp_big;
    else if (-exponent_limit > alpha_eta_a)
        exp_a = exp_sml;
    else
        exp_a = exp(alpha_eta_a);
This is a part of code for the function, `Transfer_Current`, computing transfer current, which shows all the relevant variables for the cathode. Firstly, the code for cathode-side equations were changed. There are some main variables corresponding to the Butler-Volmer equation, such as `source` for anode or cathode volumetric transfer current \( R_{\text{cat}} \); `i_ref` for effective transfer current coefficient, \( \zeta \); \( j_{\text{cat}}^{\ref} \) or \( j_{\text{an}}^{\ref} \); `alpha_c`, `alpha_a` for product of cathode or anode exchange coefficient and F/(RT): \( \alpha_{\text{cat}} F / RT \) and \( \alpha_{\text{an}} F / RT \); `pow(c_w, gamma)` for local species concentration \([C]^{\gamma_{\text{cat}}}\).

Originally, the code to describe, the cathodic Butler-Volmer equation was as:

\[
R_{\text{cat}} = \frac{\zeta_{\text{cat}} j_{\text{cat}}^{\ref} / [C]^{\gamma_{\text{cat}}} - e^{\alpha_{\text{an}} F / RT} j_{\text{an}}^{\ref} / [A]^{\gamma_{\text{an}}}}{F n \times n \times F \times [C]^{\gamma_{\text{cat}}}}
\]

This equation was amended to:

\[
R_{\text{cat}} = \left( \frac{[C]}{[C]^{\gamma_{\text{cat}}}} \right)^{-1} \left( -e^{\alpha_{\text{cat}} F / RT} + e^{\alpha_{\text{cat}}^{\ref} F / RT} \right).
\]

In the current code, `alpha_a` should not been used anymore. Instead of `alpha_a`, `alpha_c_re` is introduced. Same with `alpha_c`, `alpha_c_re` is define as \( \alpha_{\text{cat}}^{\ref} F / RT \), \( \alpha_{\text{cat}} = 1 - \alpha_{\text{cat}} \), the additional code should be:

\( alpha\_c\_re = F\_over\_R/T - alpha\_c; \) And `alpha_eta_c_re` insteded of `alpha_eta_a`, `exp_c_re` instead of `exp_a`. The code was changed to:

\[
A_p = i_{\text{ref}}^{\ast}(\exp_{c} - \exp_{a})/(\text{Farad}^{\ast}\text{n\_electrons}); /* Butler-Volmer */
\]

else

\[
A_p = i_{\text{ref}}\exp_{c}/(\text{Farad}^{\ast}\text{n\_electrons});
\]

...
### Original code

```c
if (Butler_Volmer)
{
    alpha_eta_a = alpha_a*eta;
    if (exponent_limit < alpha_eta_a)
        exp_a = exp_big;
    else if (-exponent_limit > alpha_eta_a)
        exp_a = exp_sml;
    else
        exp_a = exp(alpha_eta_a);

    A_p = \frac{i_{ref}(exp_c-exp_a)}{(Farad*n_electrons)};
}
else
    A_p = \frac{i_{ref}exp_c}{(Farad*n_electrons)};
```

### Edited code

```c
if (Butler_Volmer)
{
    alpha_eta_c_re = alpha_c_re*eta;
    if (exponent_limit < alpha_eta_c_re)
        exp_c_re = exp_big;
    else if (-exponent_limit > alpha_eta_c_re)
        exp_c_re = exp_sml;
    else
        exp_c_re = exp(alpha_eta_c_re);

    A_p = \frac{i_{ref}(exp_c-exp_c_re)}{(Farad*n_electrons)};
}
else
    A_p = \frac{i_{ref}exp_c}{(Farad*n_electrons)};
```

The new code presents the Butler-Volmer equation as:
\[ R_{cat} = \zeta_{cat}^{ref} / [C]_{ref}^{Y_{cat}} (e^{\alpha_{cat}F\eta_{cat}/RT} - e^{(1-\alpha_{cat})F\eta_{cat}/RT})/F{n\times n\times F}[C]^{Y_{cat}} \]

It has been found that the Module works well with the modified code file. The same modification of the code for the anode Butler-Volmer equation was done similar, introducing \textit{alpha\_a\_re} instead of \textit{alpha\_c}.

As mentioned, we pointed out this problem with the manual for version 14.5 to the ANSYS development team. The ANSYS team accepted this argument and changed the later version 16.2 to allow \( \alpha_{an} \), \( \alpha_{cat} \) to be set to different values for the anode and cathode. In chapters 4 and 5 of this thesis, the simulation work was done by using ANSYS fuel cell module version 14.5 with the correction to the source code detailed in section 4.2 above. Later, the simulation work reported in chapters 6 and 7 was done by using ANSYS version 16.2, which incorporated the amendment proposed in this section.

### 4.3 ANSYS FUEL CELL MODULE SIMULATION OF A BASIC PEM FUEL CELL WITH A SINGLE STRAIGHT FLOW CHANNEL FOR EACH GAS

#### 4.3.1 Introduction

To illustrate the setting up, inputs, running and outputs of the ANSYS FC Module, the basic case of a PEM fuel cell, with an active area in the shape of long rectangle or strip with a single straight gas flow channel on each side of the MEA, one for hydrogen and the other for oxygen, is presented in this section.
4.3.2 Geometry and mesh of the basic fuel cell model

Every PEM fuel cell consists of anode and cathode end plates, gas channels, gas diffusion layers, and catalysts, together with a nafion membrane. Here a model of a PEM fuel cell with a single channel on the anodic side and another single channel on the cathodic side in the ANSYS Fuel Cell Module. This simplest fuel cell model could explore the relationships between the various input parameters and the cell performance in terms of the voltage-versus-current density curve, current density distribution across the membrane, and overall energy efficiency.

The representation of this basic fuel cell in ANSYS is shown in Figure 4.5. The cell modelled is a $6\text{ mm} \times 8.79\text{ mm} \times 25\text{ mm}$ cuboid. The upper plate is the anode side plate for hydrogen. The lower plate is the cathode side for oxygen or air. The gas channel is 2 mm wide and 2 mm high on each side. The thicknesses of the gas diffusion layers, catalysts and membrane are 0.3 mm, 0.02 mm and 0.15 mm respectively. The two end plates are treated as solid objects. The rest of the parts of the fuel cell are considered as fluid objects.

![Figure 4.5: The dimensions of the basic fuel cell modelled with the ANSYS Fuel Cell Module](image)

The generation of the mesh is one of the most critical aspects of the simulation. Too many cells in the mesh may result in long solver runs, and too few may lead to inaccurate results. It is important to balance these requirements. The mesh used in this model is shown in Figure 4.6. For the end plates and the gas channels, the edges are divided up into strips 0.2 mm wide. Therefore, there are 30 cells in the horizontal dimension and 20 in the vertical dimension.
Along the length of the fuel cell, the 25 mm dimension is divided into 50 cells. The gas diffusion layers and membrane are divided into ten layers, and each catalyst layer is divided into four. The horizontal divisions of the GDLs, catalysts and membrane are the same as for the end plates.

![Image of fuel cell model](image)

Figure 4.6: The mesh of the basic fuel cell modelled

### 4.3.3 Input values for the simulation

After generation of the mesh, the description is transferred from mesh to FLUENT. The ANSYS Fuel Cell Module in FLUENT is then called up and its basic options defined as in Figure 4.7
The geometry of the single cell is then matched to the input specifications, using the dialogue box shown in Figure 4.8. Since this model of basic fuel cell, it cannot relate directly to a real experimental cell. Hence the input parameters used in the simulation are default values.
There are several zones that must be specified in the boundary conditions dialogue box. These are the anode and cathode voltage terminals, as well as the inlets and outlets to the cell. The anode side terminal is set at a potential of 0 V, and the cathode side terminal is set as 0.9 V initially. The pressure of the hydrogen inlet is set at 1 bar gauge, with a 0.99 mass fraction for H₂, and 0.01 for H₂O vapour. The pressure of the oxygen inlet is also set at 1 bar gauge, at a mass fraction of 0.8 for O₂, and 0.1 for H₂O vapour. The type of outlets are set as ‘pressure outlet’, which is a gas outlet driven by pressure.

4.3.4 Outputs of simulation

Then the default setting is used to run the simulation for 100 iteration for each overall cell voltage setting changed from 0.9 to 0.1 V at 0.1 V intervals. Based on the simulation results, the V-I curve for this single cell obtained is as shown in Figure 4.9.

![Figure 4.9: V-I curve of a single channel fuel cell](image)

In order to study the effect of different parameters on the output performance of the cell, four parameters in the input dialogue box were modified: namely the two Reference Current Densities and two Exchange Coefficients on both sides (Figure 4.10).
4.3.5 Effects of varying inputs on outputs and cell performance

In theory the exchange coefficient cannot be larger than 2. Hence the values of the exchange coefficient used were 0.5, 1.0, 1.5 and 2 for both oxygen and hydrogen, and simulations were run for each of these values again for a full set of V-J values to get the series of curves presented in Figure 4.11.
As the exchange coefficient decreases, the V-J curve is lowered (that is, a lower voltage for a given current). The slopes of the curves for exchange coefficients of 0.5 and 1 are clearly different, while those for the other values tend to similar values as J increase. Since the exchange coefficient appears as a positive multiplying factor in the exponent of the dominant term in the Butler-Volmer equation, this change in slope with decreasing exchange coefficient is to be expected. In addition, the larger the exchange coefficient, the closer the curve gets to a straight line.

Simulations were also run for a range of values for the reference current density on both side: the default values were multiplied by 0.1, 1.5 and 4. A V-J curve was obtained for each reference current density values, as presented in Figure 4.12.
As expected from the Butler-Volmer equation again, increasing the exchange current density raises the V-J curve (that is, lifts the current density for a given voltage). For the higher range of current density, all the V-J curves for the different reference current densities are approximately parallel with each other.

The reference current density and exchange coefficient used as inputs to the module can thus be changed to modify the output V-J curve in an effort to make the simulation result compare with experimental data.

As another example, the change of water content in the membrane will change its proton conductivity, which will in turn affect the output performance of the fuel cell (Figure 4.13). To illustrate this effect, the initial water content to the model was varied as follows: 0.5, 1, 1.5, 2, 3, 5, 10. When the water content increases, the membrane conductivity is increased, and the current density correspondingly increases significantly. When the current density reaches a very high value, the mass transfer limitation is present in the graph. It can be seen that the final point on the curve for a water content of 10 unexpectedly has a current density lower than the preceding point. This reduction has occurred most likely because some parameters values become out of range during the calculation. Hence the final two points are unlikely to be physically meaningful.
The limitation shows there is a maximum value for current. In a real experimental fuel cell, there is always a sharp turn down at the end of the curve that limits the maximum current. Later in the large fuel cell simulation modelling (chapter 6), the factors that affect the current constraint will be studied in more detail.

4.4 CONCLUSION

In this chapter the fundamental equations of the ANSYS Fuel Cell Module have been presented. A correction has been made to the Butler-Volmer equations in the module to allow for separate specification of the input values for the exchange coefficients on both the anode and cathode. The changes have been incorporated into the source code of the ANSYS module. The use of the ANSYS Fuel Cell Module to simulate a basic fuel cell with a single straight gas flow channel for each gas has been presented. The input parameters typically required by the ANSYS module are listed, and the usual outputs available after running the model are presented. The outputs show the influence of varying key input parameters such as reference
current density and exchange coefficient on the voltage versus current curves that are obtained from the modelling. These relationships are used in later chapters to guide the fitting of simulation results to experimental data.
5. **ANSYS FUEL CELL MODULE**

**SIMULATION OF SMALL SINGLE PEM FUEL CELLS**

5.1 **Small Single Experimental PEM Fuel Cells Investigated**

5.1.1 **Introduction of experimental fuel cells**

There are three similar small single experimental PEM fuel cells used in this thesis to compare experimental results with the ANSYS Fuel Cell Module simulation results. The purpose of the comparison is to assess how close validate fit between the module simulation and experimental results can be obtained after varying certain input parameters to the model. The three small single PEM fuel cells investigated are:

Cell A: an earlier PEM fuel cell made at RMIT University by Doddathimmaiah (2008);

Cell B: the first experiment fuel cell made in this project;

Cell C: the second experiment fuel cell made in this project
<table>
<thead>
<tr>
<th>Date</th>
<th>Geometry</th>
<th>Comments</th>
</tr>
</thead>
</table>
| Cell A | 2008 | Parallel flow channel with headers (2 mm (width)×2 mm (depth))  
Active membrane area: 5 cm² | Data obtained from previous work (Doddathimmaiah, 2008) used in the present study |
| Cell B | 2015 | Parallel flow channel with headers (2 mm (width)×2 mm (depth))  
Active membrane area: 5 cm² | New material for membrane and catalysts |
| Cell C | 2016 | Parallel flow channel with headers (1 mm (width)×1 mm (depth))  
Active membrane area: 5 cm² | Power output improved |

Table 5.1: Small single fuel cells experimented and simulated

5.1.2 Earlier PEM fuel cell made at RMIT University

The earlier PEM fuel cell made at RMIT University was designed by Doddathimmaiah (2008); this was a unitised regenerative fuel cell. This reversible fuel cell is aiming for performance in electrolyser mode and fuel cell mode in terms of energy efficiencies equal to or only slightly lower than a comparable dedicated PEM electrolyser and dedicated PEM fuel cell, and to achieve a total system cost for a URFC that is well below the total cost for a system comprising a comparable separate PEM electrolyser and PEM fuel cell. However, its operation in fuel cell mode only was considered in the present work. Experimental fuel cells with different catalyst types and loading were designed and constructed by Doddathimmaiah (2008). The MEAs used had nafion membrane with an active area of 5 cm². The design of the
experiment employed by Doddathimmaiah (2008) to measure the V-I curve for these cells is shown in Figure 5.1.

Figure 5.1: The design for PEM fuel cell A (Doddathimmaiah, 2008)

Each end plate had six parallel channels connected by headers. The dimensions of flow channels on H-side and O-side used the experiment are detailed in Figure 5.2 and Figure 5.3.
Figure 5.2: O-side flow-channel dimensions used in experimental PEM fuel cell A. (Doddathimmaiah, 2008)

Figure 5.3: H-side flow-channel dimensions used for the experimental Cell A. (Doddathimmaiah, 2008)
The experimental rig of experimental system including gas supply and measuring tools, such as gas flow meters, and multi-meters is shown in Figure 5.4

![Experimental set up](image)

Figure 5.4: Experimental set up used by Doddathimmaiah (2008)

These earlier experiments investigated the effects on V-I curves and performances of different catalyst types and loading. The combination found experimentally to have the highest energy efficiency was chosen to compare with the ANSYS Module simulation in this project in terms of V-J curves and energy efficiency.

### 5.1.3 PEM fuel cell B made in this project

PEM fuel cell B made for this project used the design of the earlier Doddathimmaiah (2008) experimental fuel cell. The geometry for the two endplate was the same as in the earlier experiment, but all the endplates and gaskets used in the present project were newly made. The two endplates were made of stainless steel and are shown in Figure 5.5.
The MEA was purchased from a commercial company (Fuelcells Etc) had an active area of 5 cm², with Pt catalyst at a loading of 4mg/cm² on the H-side and a 1:1 mixture of Pt/IrO₂ at a total loading of 4mg/cm² on the O-side.

The experimental fuel cell B is shown in Figure 5.6. As mentioned, although this fuel cell could operate as a unitised regenerative fuel cell, in this experiment it was used only in fuel cell mode.

The experimental rig was built as shown in Figure 5.7 comprising the fuel cell and three cylinders. One cylinder was used to collect oxygen; one cylinder was used to collect hydrogen; and the third cylinder which was higher is used to maintain the pressure and collect exhaust oxygen.
Figure 5.7: Experimental set up for performance measurement of cell B

The measurement tools are shown in Figure 5.8. The DC power source was used for generating hydrogen and oxygen in E-mode, prior to the operation in FC mode. The voltage and current measurements were done by two multi-meters. A programmable load was used, decrease the resistance from a very high value to a low value to increase the current from zero to its maximum value.

Figure 5.8: Measurement equipment used in the experiment with cell B
5.1.4 PEM fuel cell C made in this project

PEM fuel cell C had a number of changes to the flow channel geometry compared to cell B. The width, depth and gap between two channels were all 1 mm in cell C instead of 2 mm in cell B, aiming for a better supply of gases to the catalyst layer and hence a more even of current density. Drawings of the hydrogen plate in cell C is shown in Figure 5.9, and the oxygen plate in Figure 5.10.

![Diagram of PEM fuel cell C](image)

Figure 5.9: Hydrogen plate of experimental cell C
The active membrane area was the same as in cell B. The other components were also the same and newly purchased. The experiment with cell B was conducted in a new rig designed and constructed at RMIT University for an Australian Defence-funded project (RMIT CTD project, 2015). This rig provided a controllable environment in terms of temperature, gas flow rates, and gas relative humidities, thus allowing a more precise comparison with the ANSYS Fuel Cell Module simulation in which these variables were set to the same values as in the rig.

The rig is shown in Figure 5.11, and was fitted with separate mass flow meters, heating blocks, humidifiers, humidity meters and temperature measurement devices for both hydrogen and oxygen gases. All the tube fittings and tubing were used in this rig were obtained from Swagelok. Two separate pressure relief valves were fitted in each gas line of the rig, to ensure gas pressure at the entry of the fuel cell could not be more than 1 bar gauge.
5.2 Small PEM fuel cell simulation models conducted

5.2.1 Fuel cell model for earlier PEM fuel cell A

ANSYS Geometry was used to draw the 3D geometry of the single channel fuel cell in chapter 4. For small and large single fuel cell, it would be easier to use other software, such as Solidworks, to draw the 3D geometry. ANSYS Geometry was also used to modify the drawings to fit the ANSYS Fuel Cell Module and facilitate the meshing. This geometry included in the model covered only the area of the flow channels within an endplate and ignoring the outer area of the cell, since the latter only has a very small effect on the performance. The overall size for the cell as represented in the model was 36 mm × 36 mm × 8.7 mm. The active area of the membrane electrode assembly was 5 cm². Actual measurements found that the size for the MEA was about 22 mm × 23 mm. The catalyst layers and gas diffusion layers were built in a single product within the membrane electrode
assembly. But in the 3D representation in the model, these different layers were defined separately. Each end plate had a parallel flow channel configuration, with the width and depth of the flow channel being 2 mm × 2 mm. The modelled area only covered six channels in the middle. Figures 5.12 – 5.14 show the geometry created in ANSYS for the modelling and simulation. In order to get a more even distribution of current density, gas distribution and temperature, the width and depth of flow channel were also changed to 1 mm × 1mm as a second case for comparison with the experiments made for this project.

Figure 5.12: 3D geometry of cell A as represented in the ANSYS module

Figure 5.13: Fuel cell flow channels in the ANSYS module
There was a strip around the active membrane area between the two endplates, where the gaskets rested. The gaskets were not involved in the cell reactions and their role is just to seal the gas channels. The membrane itself was larger than the active area, reaching the edges of the endplates, in order to prevent any gas crossing from the region with the cell on one side of the membrane to the corresponding region on the other side of the cell. In the model, the area outside of the active area of the cell is not included in the simulation, and the corresponding boundaries were the gas isolation set as an ideal impenetrable wall.

During the meshing process, the ‘physics’ preference was set to ‘CFD’, and the solver preference to ‘Fluent’. The advanced size function option was not used. Edge sizing for every edge of the endplates and gas flow channels was set at 0.5mm. The length and width of each element in the membrane, catalyst and gas diffusion layer was 0.5mm. For the thickness, the catalyst layers were divided into four mesh layers, and the membrane and gas diffusion layers into six mesh layers. The total number of elements in the simulation was 147,944. Figure 5.15 presents the detail of the meshing. Because of the limited colours available, the endplate and some of the channels happened to have the same colour, but that does not mean they are the same component.

The mesh elements were all kept as hexahedra, which is preferable given that the simulation covered the calculation of gas flow as well as electrical current. It was found that when some of the mesh elements became tetrahedra with very sharp corners, it was likely to cause a
computed output that was unphysical in that area, such as an extremely high electrical potential.

![Figure 5.1: Outside and inner view of the mesh used to model the multi-channel fuel cell A in the ANSYS Module](image)

The parameters of this fuel cell previously obtained from Doddathimmaiah (2008) and material specification form companies were input to the fuel cell model as shown in Table 5.2.
<table>
<thead>
<tr>
<th>Anode</th>
<th>Unit</th>
<th>Cathode</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ref. Current Density</td>
<td>2.8 A/m²</td>
<td>Ref. Current Density</td>
<td>4.8e-6 A/m²</td>
</tr>
<tr>
<td>Ref. Concentration</td>
<td>0.04 kmol/m³</td>
<td>Ref. Concentration</td>
<td>0.03 kmol/m³</td>
</tr>
<tr>
<td>Concentration exponent</td>
<td>1</td>
<td>Concentration exponent</td>
<td>1</td>
</tr>
<tr>
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<td>Exchange Coefficient</td>
<td>0.703</td>
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<td>Porosity(GDL)</td>
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<td>Viscous Resistance(GDL)</td>
<td>1e+12 l/m²</td>
</tr>
<tr>
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<td>Porosity(CL)</td>
<td>0.285</td>
</tr>
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<td>viscous Resistance(CL)</td>
<td>1e+12 l/m²</td>
</tr>
<tr>
<td>surface/Volume Ratio</td>
<td>2e+4 l/m²</td>
<td>surface/Volume Ratio</td>
<td>2e+4 l/m²</td>
</tr>
<tr>
<td>Hydrogen inlet gauge pressure</td>
<td>10 pa</td>
<td>Oxygen inlet gauge pressure</td>
<td>10 pa</td>
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<table>
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<th>Unit</th>
<th>Overall</th>
<th>Unit</th>
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<td>Open-Circuit Voltage</td>
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<td>Protonic Conduction Coefficient</td>
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<td>Specific Leakage Current</td>
<td>0 A/m³</td>
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<tr>
<td>Protonic Conduction Exponent</td>
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<td>Electrolyte Project Area</td>
<td>5.06e-4 m²</td>
</tr>
</tbody>
</table>

Table 5.2: Input parameters in the ANSYS Fuel cell Module simulation of cell A

5.2.2 Fuel cell model for PEM fuel cell B

The design of fuel cell B was same as for fuel cell A, so the geometry input for cell B to ANSYS Fuel Cell Module model was also the same, as was the mesh. But the membrane, gas
diffusion layers and catalysts for cell B were newly purchased, and the properties of these layers were different to those in cell A as were the corresponding parameters input to the model, for reference exchange current densities, and charge transfer coefficients. The initial settings for those unknown parameters were taken from the earlier setting values for cell A in Table 5.2.

5.2.3 **Fuel cell model for PEM fuel cell C**

Since the design for fuel cell C had changed, the geometry of fuel cell for simulation was redrawn. The fuel cell model had 12 parallel channels and 2 headers with the (see Figure 5.16).

![Figure 5.16: (a) Overall view of the geometry; (b) Close view of the GDL, catalyst and membrane](image)

The end plates were made from stainless steel and are 3mm in thickness. The gas diffusion layer, catalyst and membrane were bought from commercial suppliers. The thickness used for the gas diffusion layer was 0.43mm; the thickness of catalyst and membrane together was 0.14 mm. By splitting the catalyst and membrane, the thickness of the membrane used was 0.1 mm.

The overall meshing used in modelling the fuel cell in the ANSYS module is shown in Figure 5.17. The meshing used in the module for the various layers is shown in Figure 5.17. It can be seen that in some layers, for example the catalyst layers, the mesh need to be much fine
because of the variation in key parameters that can occur over relatively short distances in these layers.

In the ANSYS fuel cell module, the voltage of the cell was first provided for calculation of the performance, including cell current. For the base case, the V-I curve should cover the whole range of the theoretical performance. As the open circuit voltage was 1.2 V (higher potential is on cathode side). The actual starting working voltage was lower than 1.2 V, actually 1.0 V, and the simulation was run for 10 voltages each falling by 0.1V.

5.3 Small PEM fuel cell models: simulation results

5.3.1 Cell A

The ANSYS Module simulation for cell A was run by setting a cell voltage and then calculating the current flow throughout the cell as. The input cell voltages were from 0.8 V down to 0.1 V in 0.1 V steps. In this project, average current density was used instead of total current in the Voltage versus current curve, so that it was really a voltage versus current density (V-J) curve, which allows meaningful comparisons with other fuel cells with different
membrane active areas to be made. The simulation result of cell A using ANSYS Module is shown in Figure 5.18.

![V-J Curve](image)

**Figure 5.18: V-J Curve of cell A using ANSYS Fuel Cell Module**

Examples of other outputs from the ANSYS Module simulation for cell A are provided here for a cell voltage set at 0.7 V and the temperature at 300 K. The calculation ran for 200 iterations. Figures 5.19 and 5.20 show the mass fraction and velocity magnitude in the hydrogen flow channel respectively. From Figure 5.19, the decrease in mass fraction of hydrogen occurs progressively along the channels as the reaction of hydrogen molecules in the catalyst layer takes place, leading to their splitting into H⁺ ions and electrons.
Figure 5.19: Simulated mass fraction of hydrogen in the gas flow channel in cell A

The velocity of hydrogen map (Figure 5.20) shows the largest hydrogen flows through the outer channel. Lower hydrogen flow velocity occurred in the central channels of the cell.

Figure 5.20: Simulated velocity of hydrogen in the channels

Current flux density at the surface between the anode catalyst layer and gas diffusion layer, is shown in Figure 5.21. The edges of each of six channels have higher current density than other parts. It can tell that the current density distribution is not even enough, so the reaction
was mainly happened on the edge of the channels. The reaction needs the gas channels to provide gas and the ribs to provide the electrical pathway.

![Current Flux Density in the membrane of cell A (0.3V)]

Figure 5.21: Current Flux Density in the membrane of cell A (0.3V)

The current density pattern in Figure 5.21 suggests that the width of the channel and the distance between two channels were too large, because there are significant drops in current density between channels, and in the middle of the channels (Figure 5.22). The Figure 5.22 shows the current flux density in the membrane in the direction as cross the channels. The peak came at the edge of the channel. There is a low point between two peaks in one channel, because the channels are not conductors, the current cannot go through.
The optimal channel configuration should consider the gas flow as well as the electrical current flow. Although wide channels enhance gas supply, but at the same time less area on the ribs for electrical contact with the GDLs have an adverse effect on cell resistance. For most commercial fuel cells, the width of gas channels is less than 1 mm.

The simulated V-J curve for cell A is compared with the corresponding curve for cell A obtained by Doddathimmaiah (2008) in Figure 5.23. A voltage versus current (V-I) curve of the earlier PEM fuel cell was obtained from report.

Figure 5.22: Current flux density magnitude in the membrane

Figure 5.23: V-J curve form ANSYS simulation result and the experimental curve
The curve of ANSYS model followed the trends of the experiment’s curve. The maximum current densities were quite close, 0.121A/cm\(^2\) at 0.095V for experiment, 0.124A/cm\(^2\) at 0.1V for ANSYS model. And starting points were similar too, 0A/cm\(^2\) at 0.754V for experiment, and 0A/cm\(^2\) at 0.8V for ANSYS model. In the middle range of voltage, the current densities of ANSYS model were larger than the experiment. For comparing the difference, calculate the current density of experiment at each voltage point using in ANSYS model (Table 5.3). At 0.4V, the current density of ANSYS model was higher than the experiment by 28% of the experimental current density. The ANSYS Fuel Cell Module presented the result in the reasonable range of the real fuel cell.
5.3.2 Cell B

5.3.2.1 Measuring experimental performance

The multi-meters were used to measure current and voltage for cell B over a range of values. Current was converted to current density to obtain the V-J curves shown in Figure 5.24. Data sets for three experimental runs were obtained with the first run giving the best performance and a maximum current density 0.1 A/cm². It is likely that there was a build-up of liquid water in the cell after successive runs with partial blockage of the pores in the gas diffusion and catalyst on the oxygen side limiting oxygen supply to the catalysts. At the end of the experiment when the cell was opened, lots water was found in the MEA. This is probably the explanation of why the maximum current density was low for a PEM fuel cell even in the first run.
In the following chapter, the issue of excessive water accumulation will be discussed again, since a similar problem occurred in the later experiments.

For matching with the output of the ANSYS Module simulation for cell B, the V-J curve obtained in the first experiment run was used.

5.3.2.2 Varying inputs to simulation to get best fit to experimental data

To obtain the best fit between the simulation and experimental V-J curve of cell B, the reference exchange current densities and exchange coefficients on both electrodes, and the initial water content in the membrane (that is, the number of water molecules sorbed per sulfonate group) were varied as the inputs to the simulation. The three sets of input values are presented in Table 5.4.
The ANSYS V-J curves obtained for these three models are shown in Figure 5.25 along with the corresponding experimental curve. Because of the limitation of computing time available, limited points were obtained for each ANSYS model.

**Table 5.4: The three sets of input parameters used in the simulation models of cell B**

<table>
<thead>
<tr>
<th>Model</th>
<th>Ref. current density for anode (A/m²)</th>
<th>Ref. current density for cathode (A/m²)</th>
<th>Exchange coefficient for anode</th>
<th>Exchange coefficient for cathode</th>
<th>Initial water content (λ)</th>
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<tbody>
<tr>
<td>Model 1</td>
<td>0.0028</td>
<td>9.00E-07</td>
<td>1.7</td>
<td>1.3</td>
<td>3.3</td>
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<td>Model 2</td>
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<td>1.3</td>
<td>5</td>
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<tr>
<td>Model 3</td>
<td>0.0028</td>
<td>4.78E-07</td>
<td>1.7</td>
<td>1.3</td>
<td>3.3</td>
</tr>
</tbody>
</table>

**V-J Curves between Modelling and Experiment data**

Figure 5.25: The V-J curves obtained from the ANSYS Module simulation compared with the experimental data

**Compare the curves using variance.**

\[
\text{Var}(x) = \frac{\sum_i\left[I_{\text{seti}}(V_x) - I_{\text{experiment}}(V_x)\right]^2}{x}
\]
<table>
<thead>
<tr>
<th>Voltage (V)</th>
<th>Experiment Current Density (A)</th>
<th>ANSYS Model 1 Current Density (A)</th>
<th>ANSYS Model 2 Current Density (A)</th>
<th>ANSYS Model 3 Current Density (A)</th>
</tr>
</thead>
<tbody>
<tr>
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<td>0.000190788</td>
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</tbody>
</table>

Table 5.5: Variance between experiment data and ANSYS models

5.3.2.3 Best-fit input values

From Table 5.3, it is clear that model 3 gave the smallest variance between the simulated V-J data and the experimental results. The closeness of the fit is shown in Figure 5.26.

![V-I Curves between Modelling and Experiment data](image_url)

Figure 5.26: Fitted ANSYS Model and experiment data
5.3.3 Cell C

5.3.3.1 Measuring experimental performance of cell C

Different form cell B, cell C was run with air instead of pure oxygen. The experimentally-measured V-J curve is shown in Figure 5.27. Air was allowed to flow through the cell with the outlet partly open, while the hydrogen side was dead-ended with regular purging. The maximum current density of 0.385 A/cm² was obtained at potential of 0.37V. The maximum power output for cell C occurred around 0.7V.

The performance of the experiment cell C gave quite good curve of output performance. The maximum current density was 0.37 A/cm², while the maximum current density was 0.12 A/cm² for cell A and 0.1 A/cm² for cell B. The result suggests that the changing of geometry of cell C was improving the performance.

Figure 5.27: The V-J curve for fuel cell measured by experiment
5.3.3.2 Varying inputs to simulation to get best fit to experimental data

Some of the setting value may not be the same with cell B. In case of same current output, the amount of gas fit in should be about five times, since oxygen only takes about 20% of air. In fact, the model is allowed the gas to flow through the channel, and the conditions of the gas in flow channel will also change. The input mass flow rate of gas started with the same value as used for cell B, and then it was changed to get a reasonable output close to the experiment.

The operating voltage points in the experiment were different from those in the computer simulation, since fuel cell C’s performance was very sensitive to the load and it was thus very hard to set the output to a predetermined repeatable voltage. Hence the points on the experimental V-J curve corresponding to the voltage settings in the simulation were obtained by interpolation on the experimental curve as shown in Figure 5.28.

![Experimental V-J curve for cell C](image)

**Figure 5.28:** Interpolation on the experimental V-J curve for cell C to obtain the experimental points corresponding to the voltage values used in the simulation

The ANSYS simulation model was modified to reflect the same geometry of cell C. The initial settings of fuel cell parameters in the simulation were the same as those for cell B. The
air-side flow rate was started from five times the oxygen flow rate used in cell B. The V-J curve generated by the simulation is shown in Figure 5.29 using of the same input parameters as for best-fitted simulation of cell B.

![V-J curve of experiment and simulation](image)

Figure 5.29: The curves of voltage vs. current density of experiment and initial input values as for cell B simulation

The simulation result had a higher voltage at the beginning of the curve, while the two curves overlapped from 0.05 A/cm² to 0.13 A/cm². The experiment curve had a higher voltage to 0.34 A/cm², and then drop below the simulation curve.

The spatial variation of the current density distributions at 0.7 V across various planes in the cell generated by simulation are shown in Figure 5.30.
Figure 5.30: Simulated current density distributions for cell C: (a) hydrogen-side end plane and channel, (b) hydrogen-side gas diffusion layer, (c) hydrogen-side catalyst, (d) central plane of membrane, (e) oxygen-side catalyst, (f) oxygen-side gas diffusion layer, and (g) oxygen-side end plate and channel.

From these current density distributions, it can be seen that the current density in areas aligned with the ribs is generally higher than that in areas aligned with the channels, reflecting the obvious property that the gas channels will not allow current to go through them. The current density in the central plane of the membrane (Figure 4(d)) is in the range 1.10x10^4 A/m^2 to 1.16x10^4 A/m^2, which is not a large variation. The result thus shows that at a cell voltage of 0.7 V, the current distribution in the membrane is quite even. Hence the design of the flow channel configuration and geometry of the cell is acceptable.

The graphs in Figure 5.31 show the variations in spatial distribution of current density in the middle of the membrane between the voltages 0.9V and 0.4V. The range of current density in each graph is relatively narrow, with the highest value only about 4% higher than the lowest.
value. Hence these distributions can be considered relatively even, as required for a well-functioning fuel cell. But there were still some differences in the same graph; the highest current density points came at two top corners. Figure 5.31 (a) showed a different pattern from other figures, since the current density is relatively lower than in the other graph. The current distribution patterns at higher current points showed similar features.

Figure 5.31: Effect of cell voltage on spatial distribution of current density in the middle of the membrane.

5.3.3.3 Best-fit input values

The fitting process of the simulated to the experimental curves used for cell C was the same as that for cell B. The reference exchange current density and exchange coefficients used as inputs to the ANSYS simulation were changed to modify output of the model. Some sets of input parameters were tested in the fuel cell model. The various V-J curves obtained from the simulation model for these sets of input values are compared with the experimental curve. Three simulation curves most close to the experimental curve was obtained in Figure 5.32.
The coordinate values for the selected cell voltages taken from the curves in Figure 5.31 are listed in Table 5.6.

<table>
<thead>
<tr>
<th>Voltage (V)</th>
<th>Current density (A/cm²)</th>
<th>experiment</th>
<th>set1</th>
<th>set2</th>
<th>set3</th>
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<tr>
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</table>

Table 5.6: Current densities from the ANSYS simulations corresponding to the selected cell voltage using the three sets of input parameters, along with the respective experimental current densities
Compare the output of three sets of input value by calculating the variance between simulation result and experiment. As equation,

\[
\text{Var}(x) = \frac{\sum_i [I_{\text{set}_i}(V_x) - I_{\text{experiment}}(V_x)]^2}{x}
\]

<table>
<thead>
<tr>
<th>Voltage</th>
<th>set1</th>
<th>set2</th>
<th>set3</th>
</tr>
</thead>
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</tbody>
</table>

Table 5.7: Variance between experiment data and ANSYS models

From Table 5.7, it can be seen that, when voltage was higher than 0.6V, set 3 gave the closest values to the experimental results. However, none of the simulated curves was able to reflect the sharper turn-down in voltage at high current density shown in the experimental curve. The important question of how to get V-J curves generated by the ANSYS Fuel Cell Module to represent more accurately this limitation of current density will be addressed in detail in chapter 6.3.

Further work would be done to simulate the limitation effect at higher current situation. The input setting of set 3 could be suitable for the simulation of the larger fuel cell modelling. The full settings of set 3 shows in the following table including all the input parameters.
### Table 5.8: Major input parameter for best fitted simulation of cell C

<table>
<thead>
<tr>
<th>Anode</th>
<th>Unit</th>
<th>Cathode</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ref. Current Density</td>
<td>100 A/m²</td>
<td>Ref. Current Density</td>
<td>10 A/m²</td>
</tr>
<tr>
<td>Ref. Concentration</td>
<td>0.04 kmol/m³</td>
<td>Ref. Concentration</td>
<td>0.03 kmol/m³</td>
</tr>
<tr>
<td>Concentration exponent</td>
<td>1</td>
<td>Concentration exponent</td>
<td>1</td>
</tr>
<tr>
<td>Exchange Coefficient (a)</td>
<td>1.9</td>
<td>Exchange Coefficient (a)</td>
<td>0.1</td>
</tr>
<tr>
<td>Exchange Coefficient (c)</td>
<td>0.1</td>
<td>Exchange Coefficient (c)</td>
<td>1.9</td>
</tr>
<tr>
<td>Porosity(GDL)</td>
<td>0.63</td>
<td>Porosity(GDL)</td>
<td>0.63</td>
</tr>
<tr>
<td>Viscous Resistance(GDL)</td>
<td>1e+13 1/m²</td>
<td>Viscous Resistance(GDL)</td>
<td>1e+13 1/m²</td>
</tr>
<tr>
<td>Porosity(CL)</td>
<td>0.28</td>
<td>Porosity(CL)</td>
<td>0.28</td>
</tr>
<tr>
<td>viscous Resistance(CL)</td>
<td>1e+13 1/m²</td>
<td>viscous Resistance(CL)</td>
<td>1e+13 1/m²</td>
</tr>
<tr>
<td>surface/Volume Ratio</td>
<td>2e+5 1/m²</td>
<td>surface/Volume Ratio</td>
<td>2e+5 1/m²</td>
</tr>
<tr>
<td>Hydrogen inlet Mass flow rate</td>
<td>6e-7 kg/s</td>
<td>Oxygen inlet Mass flow rate</td>
<td>7e-6 kg/s</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Membrane</th>
<th>Unit</th>
<th>Overall</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Equivalent Weight</td>
<td>1100 kg/kmol</td>
<td>Open-Circuit Voltage</td>
<td>1 V</td>
</tr>
<tr>
<td>Protonic Conduction Coefficient</td>
<td>0.6</td>
<td>Specific Leakage Current</td>
<td>0 A/m³</td>
</tr>
<tr>
<td>Protonic Conduction Exponent</td>
<td>0.5</td>
<td>Electrolyte Project Area</td>
<td>5.06 cm²</td>
</tr>
</tbody>
</table>

**5.4 Conclusions**

In this chapter, the V-J curves generated by the ANSYS Fuel Cell Module for three cells for which experimental V-J curves were also available are compared. Best-fit values for key input parameters, including principally reference exchange current densities and exchange coefficient, to the simulation were obtained by minimising the variance between the simulated and experimental curves.
Fuel cell C in particular produced an acceptable voltage versus current density curve. The curve dropped quickly at the beginning, then decreased slowly with constant rate, and in the end the rate of fall was increasing. At this stage, the turndown of the curve at the end was not considered in the model. The fitting process focused solely on the range before the maximum output power. The best-fit inputs for the simulation of fuel cell C were later used as inputs for the ANSYS Fuel Cell Module simulations of a much larger fuel cell and fuel cell stacks. The issue of the turndown of the end part of V-J curve is covered in chapter 6.
6. SIMULATION OF A LARGE PEM FUEL CELL

6.1 INTRODUCTION

As the experimental testing and simulation of the small fuel cells in the previous chapter has shown, the power output was only around 1W. Clearly large power outputs per cell are required for most practical applications. Increased power from a fuel cell can be obtained both by increasing the active area of the cell, which raises the current for the same cell voltage; and/or by stacking individual cells in series, which increases the voltage while keeping the current the same. In the present chapter, a single PEM fuel cell with a relatively large square active area of 225 cm² (15 cm×15 cm) is simulated using the ANSYS Fuel Cell Module. The V-J curve output from the simulation is then compared with the experimentally-measured performance of the cell.

6.2 ANSYS FUEL CELL MODULE SIMULATION OF THE SINGLE LARGE CELL

6.2.1 Geometry of single large fuel cell

The large fuel cell modelled had an active membrane area (15 cm×15 cm) 45 times larger than that (5 cm²) of the small cells considered in chapter 5. The thickness of the endplates was 5 cm.
The flow channel configuration of the large fuel cell studied here was designed as part of a larger project to design and construct a 2 kW reversible fuel cell stack (RMIT University CTD Project, 2017; Sajid Mohammad, 2017). The hydrogen-side channel configuration was similar to that of the earlier small cells investigated, with the gas inlet at one top corner and the outlet at the opposite bottom corner, and parallel riser channels between the two headers (Figure 6.1a).

However, the riser channels were divided into four sections, while sharing the same header at top and bottom. The oxygen/air-side channel configuration also had the riser channels divided into four sections, but there were two gas inlets each connected to a header serving two sections of risers, and four outlets each with its own separate header (Figure 6.1 b). Overall the channel configurations on both sides of the cell occupied an area 15 cm × 15 cm.

These flow channel configurations were designed to distribute hydrogen gas and air as evenly as possible on the two sides of the cell.

The flow channel configuration as modelled in the ANSYS Module is shown in Figure 6.2.
6.2.2 Mesh for simulation

This full-sized fuel cell had a large membrane active area. The flow channels were only 1 mm wide with 1 mm ribs in between. Hence the mesh used in the simulation model had to be very small compared with the overall size, especially in the membrane active area. Figure 6.3 shows the mesh employed for view over the endplate.
Figure 6.3 and 6.4 shows the mesh at the endplate. Figure 6.3 shows the area of the endplate was divided into several parts. The top and bottom parts had through hole in the area. Figure 6.4 shows the mesh in the middle part of the endplate. The maximum size of the mesh was 0.1 mm throughout the fuel cell. The Module calculated not only the gas flow but also the electrical current. The basic meshing method was similar to the small cell, to keep the mesh hexahedra.

Figure 6.5: Mesh in gas diffusion layer, catalyst and membrane.

Figure 6.5 shows a close view of the mesh in a plane normal to the main plane of the membrane, the catalyst layers and the gas diffusion layers. The catalyst layers were formed by coating a carbon cloth, which in turn was attached to the nafion membrane. Hence these
layers were very thin, typically 12 microns thick. In the module, the catalyst layers were thus also very thin layers, with very fine mesh element dimensions (normal to membrane area) down to 1 micron. These thin layers, however, play a very important role in the electrochemical reactions. In the ANSYS Fuel Cell Module it is assumed that these reactions take place entirely within the catalyst layers. In order to get a smooth solution to the equation sin the model across the boundary between catalyst and other layers (namely the membrane and GDLs), the mesh in the neighbouring layer has to have a very low thickness like that in the catalyst. Since the active area of the cell has large lateral dimensions compared to the thickness of the various layers in the cell, the mesh elements also have lateral dimension much greater than the thickness. The minimum thickness of mesh in the endplate was 0.1 mm, while the mesh in the catalyst layers was 3e-4 mm thick.

6.2.3 Simplification of geometry

The actual cell had through holes outside of the active area of the MEA for gas delivery and removal. In order to simplify the model, its scope was reduced to only cover the area to 3 mm outside the channel area (equivalent to the membrane active area), which meant that the through holes were excluded. It was assumed that this simplification would not affect the modelled performance of the cell, since the membrane active area was still the same.

6.2.4 Input settings

The small 5 cm² PEM fuel cell model in chapter 5 had been simulated and compared with the experimental data. The best-fit values of the unmeasurable input cell parameters were found by comparing the simulated V-J curve with the corresponding experimental curve. The values of these parameters were also used as input to the model of the large fuel cell, since all materials for the MEA and GDLs were the same as for the small cell. Specifically, the reference current densities and exchange coefficients for the electrodes are material-based parameters.
The geometry of the large experimental fuel cell with 225 cm$^2$ active membrane area is shown in Figure 6.2. However, the model included the oxygen endplate, hydrogen endplate, catalysts, gas diffusion layers and membrane, but did not include the gas fittings, connections and holders.

All the principal inputs to the model are presented in Table 6.1

<table>
<thead>
<tr>
<th>Anode</th>
<th>Unit</th>
<th>Cathode</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ref. Current Density</td>
<td>100 A/m$^2$</td>
<td>Ref. Current Density</td>
<td>10 A/m$^2$</td>
</tr>
<tr>
<td>Ref. Concentration</td>
<td>0.04 kmol/m$^3$</td>
<td>Ref. Concentration</td>
<td>0.03 kmol/m$^3$</td>
</tr>
<tr>
<td>Concentration exponent</td>
<td>1</td>
<td>Concentration exponent</td>
<td>1</td>
</tr>
<tr>
<td>Exchange Coefficient (a)</td>
<td>1.9</td>
<td>Exchange Coefficient (a)</td>
<td>0.1</td>
</tr>
<tr>
<td>Exchange Coefficient (c)</td>
<td>0.1</td>
<td>Exchange Coefficient (c)</td>
<td>1.9</td>
</tr>
<tr>
<td>Porosity(GDL)</td>
<td>0.63</td>
<td>Porosity(GDL)</td>
<td>0.63</td>
</tr>
<tr>
<td>Viscous Resistance(GDL)</td>
<td>1E+12 l/m$^2$</td>
<td>Viscous Resistance(GDL)</td>
<td>1E+12 l/m$^2$</td>
</tr>
<tr>
<td>Porosity(CL)</td>
<td>0.28</td>
<td>Porosity(CL)</td>
<td>0.28</td>
</tr>
<tr>
<td>viscous Resistance(CL)</td>
<td>1E+12 l/m$^2$</td>
<td>viscous Resistance(CL)</td>
<td>1E+12 l/m$^2$</td>
</tr>
<tr>
<td>surface/Volume Ratio</td>
<td>20000 l/m$^2$</td>
<td>surface/Volume Ratio</td>
<td>20000 l/m$^2$</td>
</tr>
<tr>
<td>Hydrogen inlet</td>
<td>kg/s</td>
<td>Oxygen inlet</td>
<td>kg/s</td>
</tr>
<tr>
<td>Membrane</td>
<td>Unit</td>
<td>Overall</td>
<td>Unit</td>
</tr>
<tr>
<td>Equivalent Weight</td>
<td>1100 kg/kmol</td>
<td>Open-Circuit Voltage</td>
<td>* V</td>
</tr>
<tr>
<td>Protonic Conduction Coefficient</td>
<td>0.6</td>
<td>Specific Leakage Current</td>
<td>0 A/m$^3$</td>
</tr>
<tr>
<td>Protonic Conduction Exponent</td>
<td>0.5</td>
<td>Electrolyte Project Area</td>
<td>0.0225 m$^2$</td>
</tr>
</tbody>
</table>

Table 6.1: Major input parameter for simulation of large fuel cell

### 6.2.5 Results and analysis

The current output of the large fuel cell model was 30.83 A at 0.7 V, obtained by integrating the current density over the surface of an endplate. The power at this operating point, which was near the maximum power point, can be calculated as 30.83×0.7=21.58 W.
Figure 6.6: Current density in the membrane for the large PEM fuel cell modelled

Figure 6.6 shows the distribution of current density over the central plane of the membrane. The maximum current density was 0.211 A/cm² in the middle to bottom part of the active area (red in Figure 6.6), the minimum current density is 0.11 A/cm² at the ribs between divided four sections in the bottom. Hence the maximum current density was about eight times as much as the minimum. However, the region of middle current density, that is, from 0.125 A/cm² to 0.166 A/cm² covered about half of the total active area of the cell. Overall, the current density is generally increasing from top to bottom, from 0.110 A/cm² to 0.211 A/cm². The average current density across the full active area of the cell was 0.137 A/cm².

The distribution of water content is shown respectively in Figure 6.7.
The regions of lower membrane conductivity correspond to the areas with relatively low water content. This relationship is in line with the equations in the ANSYS Fuel Cell Module that have membrane conductivity dependent on water content in a monotonically increasing manner. It can be seen from Figure 6.7 that the top part of the cell is dryer than the lower part, leading to lower conductivity in this region, and hence lower current density. The variation in the water content across the membrane could thus be a major cause of the uneven distribution in current density.

This may due to the humidity of the air, which is not high enough to provide enough water to the membrane. When the reaction occurs along the channel, water is generated to hydrate the membrane. There are also three lowest point near the bottom. These lowest point may due to the lack of oxygen supply. Gas flow performance is shown in Figure 6.8 and 6.9.
Figure 6.8: Gas flow in the hydrogen channel (a) Velocity of hydrogen (b) total pressure of hydrogen (c) Mass fraction of hydrogen (d) Molar concentration of hydrogen
Figure 6.9: Gas flow in the oxygen channel (a) Velocity of oxygen (b) Total pressure of oxygen (c) Mass fraction of oxygen (d) Molar concentration of oxygen

From Figure 6.8 and 6.9, the molar concentration of hydrogen has a good distribution, but in oxygen channel, the molar concentration of oxygen has significant decrease along the channel. The distribution of the molar concentration may lead to the shape of current density pattern in Figure 6.6.
6.2.6 Comparison with experimental data

An experimental V-J curve was also obtained for the large fuel cell modelled in the ANSYS Fuel Cell Module and is shown in Figure 6.10 (blue curve). This experiment was conducted as part of the RMIT CTD Project (2016) with the assistance of a number of researchers working on this project, in addition to my contribution. As shown in the curve obtained, there were some problems in getting a steady increase in current density and power output, leading to voltages for current densities between 0.03 and 0.12 A/cm² lower than expected on the basis of later cell performance, when operating conditions (in particular air flow rate) were better optimised.

![Experimental vs fitted V-j curves](image)

Figure 6.10: V-J curves of Experimental data and simulation result

In Figure 6.10, the blue line is the data for the experiment. As the experimental curve shows, there was a sharp drop after 0.8V. When the current reached 0.12A/cm², the voltage dropped to 0.44 V. The experimental fuel cell was then treated by purging to remove the water and increasing the air flow rate. The voltage then increased to 0.7V while the current density remained the same. There was a second similar adjustment made when current reached 0.18A/cm².
In order to obtain a full V-J curve that corresponded to near optimal operating conditions of the cell, a smoothed curve (red line and points) was calculated, with shared beginning and end points with the blue experimental V-J curve.

The green line is the ANSYS simulation result for the V-J curve of the large cell. The simulated V-J curve fitted the smoothed experimental curve closely for just over the first half of the total range of current density. However, the experimental V-J curve fell increasingly below the simulated curve as current density approached the higher values.

Compare the variance by using the equation

$$\text{Var}(x) = \frac{\sum_i [I_{seti}(V_x) - I_{experiment}(V_x)]^2}{x}$$

<table>
<thead>
<tr>
<th>Voltage (V)</th>
<th>Current Density of fitted experimental curve (A/cm²)</th>
<th>Current Density of the simulation curve (A/cm²)</th>
<th>Variance</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.9</td>
<td>0.005</td>
<td>0.014</td>
<td>8.10E-05</td>
</tr>
<tr>
<td>0.8</td>
<td>0.053</td>
<td>0.06</td>
<td>6.50E-05</td>
</tr>
<tr>
<td>0.7</td>
<td>0.13</td>
<td>0.137</td>
<td>5.97E-05</td>
</tr>
<tr>
<td>0.6</td>
<td>0.2</td>
<td>0.31</td>
<td>3.07E-03</td>
</tr>
</tbody>
</table>

Table 6.2: Comparison of current density between fitted experimental curve and simulation curve

As Table 6.2 shows, the variances of current density from 0.9 V to 0.7 V are very small. Until the voltage drops to 0.6 V, there is a big difference of current density between fitted experimental curve and simulation curve. The curve of the fitted experiment is showing a turndown trend.

The turndown of the actual experimental V-J curve for this cell was due to the well-known mass transfer constraints that limit the reaction rates in PEM fuel cells as their maximum current density is approached (LARMINIE AND DICKS 2003). The possible ways of including the modelling of this mass transfer constraint into the ANSYS Fuel Cell Module are discussed in the next section.
6.3 Modelling the mass transfer constraint in a PEM fuel cell

6.3.1 ANSYS simulation of mass transfer constraint for large single PEM cell

In the experimental V-J curve (Figure 6.9), when the fuel cell worked at higher current and power output, the rate of increase in current density with fall in voltage declined progressively after a certain point (around \( J = 0.12 \text{ A/cm}^2 \)). Thus the V-J curve turned downwards at the end, which is known to be associated with mass transfer constraints (Larminie & Dicks, 2003). Such constraints arise due to limitations in the rate of reactants reaching the catalysts, which can be caused by many factors, including:

- the accumulation of liquid water droplets in the air-side gas diffusion layer and/or flow channels that restrict the inflow of oxygen,
- water build-up in the air-side catalyst layer that prevents oxygen molecules reaching the surface of catalyst particles,
- the limitations on the mass flow rate of input gases (usually air/oxygen) due to supply factors, channel geometry, and GDL characteristics.

In the prior ANSYS fuel cell simulation result for the V-J curve for this cell, there was no sign of this turning down of the curve at an increasing rate as the current density increased in the higher-current region up the point \([0.31 \text{ A/cm}^2, 0.6V]\) (green curve in Figure 6.10). In this part, the simulation was done below 0.6V to 0.3V, where there came a sign showing the turn down of the curve.

However, when the ANSYS fuel cell simulation was done again using the same input parameters as before but plotting points beyond 0.6 V down to 0.3 V, the turndown of the V-J curve at the high current end was clearly visible (Figure 6.11). From the point \([0.4 \text{ A/cm}^2, 0.5V]\) onwards, there was a sharp increase in the rate of voltage drop with increasing current. Hence the ANSYS simulation correctly displayed the same mass transfer constraint as the experimental V-J curve had done, but its onset was at a much higher current density: 0.4 A/cm\(^2\) rather than 0.25A/cm\(^2\) in the experiment.
6.3.2 How the mass transfer constraint is represented in the ANSYS Fuel Cell Module

To explore the reasons for the much later onset of the mass-transfer constraints in the ANSYS Module simulation as current density is increased, it is necessary to understand how these constraints are represented in the Module. The Butler-Volmer equations used in the Module to calculate current generation from the overpotentials on each electrode were described in chapter 4 (equations 4.4 in section 4.1).

It can firstly be noted that the current density is proportional to the reactant gas concentration at the electrode raised to the power ‘γ’, called the ‘concentration exponent’. In the present simulation, the concentration exponents on both electrodes were given the value of 1.0, so current density varied linearly with gas concentration. Hence one factor that can lead to a turndown in the V-J curve is reduced gas concentration as current density increases.
Secondly, the factors in the Module that influence the flow rates of reactant gases to the catalyst layers, particularly on the oxygen side, need to be considered. Two such factors are the porosities of the gas diffusion layer and the catalyst layer. In the ANSYS module, these porosities are modified by multiplying by the factor \([1-s]\), where \(s\) is the volume fraction of liquid water in the respective layer. Hence as liquid water builds up, \(s\) increases and the porosity is accordingly reduced. As porosity falls, the rate of diffusion of the reactant gas through the layer also decreases. The concentration of reactant gas reaching the catalyst layer would thus also fall, and contribute to the mass transfer constraint.

Thirdly, as the amount of liquid water in the catalyst layer increases, some of this water gathers on the surface of catalyst particles and blocks access by oxygen molecules. To take account of this effect, the specific active area in the catalyst layer is also multiplied \([1-s]\). Thus, as the volume fraction of liquid water increases, the active catalytic area is reduced accordingly, and the rate of the electrochemical reaction falls.

In order to study the limitation of current density within the ANSYS Module by water saturation (expressed as volume fraction of liquid water) and insufficient gas concentration, the spatial distributions of water saturation and gas concentration generated at the boundary between the catalyst and gas diffusion layer on the oxygen side were recorded at various voltages between 0.7 and 0.3 V.

The corresponding maps generated by the Module for the volume fraction of liquid water at this surface are presented in Figure 6.12. The scale for the volume fraction of liquid water is 0 to 1, with 0 representing no liquid water in the particular small volume (all water being in vapour form), and 1 representing complete flooding of the small volume with water. So in the blue regions of the maps in Figure 6.12 the fraction of liquid water is negligibly small, less than \(10^{-4}\) even in (e); while in the red regions the highest the fraction gets is just over 0.2. The corresponding maps of the oxygen concentration across the same surface are shown in Figure 6.13.
Figure 6.12: Volume fraction of liquid water at the boundary surface between the oxygen catalyst layer and the oxygen-side GDL at different cell voltages
Figure 6.13 Oxygen concentration at oxygen catalyst surface (touching the GDL) at different voltage

At 0.7V (Figure 6.12(a)), the amount of liquid water across this surface is very small, except for one small region in the lower middle of the active area, where its volume fraction rises to
about 0.2. Interestingly this point corresponds to a minimum in oxygen gas concentration shown in Figure 6.13(a). At 0.6 V (6.12(b) water saturation rises in three lower regions but only to ~0.06, again corresponding to regions of lower oxygen concentration in 6.13(b). At 0.5 V and increasingly at 0.4 V, the regions of higher water saturation (0.07 to 0.09) spread to an increasing fraction of the active area, and are associated with similar regions of lower oxygen concentration. Finally at 0.3 V (6.12(e)), the regions of relatively high water saturation (over 0.1) have spread to around 80% of the whole active area, while a similar region shows signs of oxygen starvation (6.13(e)).

Hence from these maps there are clear indications of the expected close linkage between increasing volume fraction of liquid water – that is, water saturation – as cell voltage declines and current density increases, and declining concentration of oxygen gas.

The average volume fraction of liquid water across the whole active area is plotted against the corresponding average current density, along with the usual V-J curve, in Figure 6.14. The water saturation increases with current density from about 0.13 A/cm², with increasing becoming ever more rapid at higher J. There is thus a clear indication that the liquid water could not be removed efficiently at the highest current densities, leading to reduced oxygen concentration and a corresponding limitation in cell output.

The following figures show the water saturation and gas concentrations at different current density. The large fuel cell module was using a constant air gas supply, so the gas concentration would decrease because of the consumption of reactant gas. In fully functional fuel cell, the air supply would increase to keep the gas concentration.
Further evidence for this explanation is provided by the graph of average oxygen concentration across the active area of the cell versus average current density, along with the corresponding V-J curve, plotted in Figure 6.15. The oxygen concentration drops steadily with increasing average current density up to 0.3 A/cm$^2$, and then with further increases in current density the steep decline in oxygen concentration mirrors the similar sharp drops in cell voltage.
The graph of average hydrogen concentration versus average current density (with all averages taken over the active area) based on the ANSYS Module simulation is significantly different from that on the oxygen side (Figure 6.15). The hydrogen-side plot does not show the sharp drop in hydrogen concentration as the cell voltage drops at high current density. By contrast the hydrogen concentration stays at about the same value with rising current density at the end of the curve. By comparing the oxygen and hydrogen concentrations, the hydrogen concentration dropped by only 10% in total over the full range of current densities, while the oxygen concentration dropped by about 90% from beginning to end.
Taken together, the gas concentration plots on both the oxygen and hydrogen sides suggest that, while declining oxygen concentration in the oxygen-side catalyst layer played a key role in limiting the maximum current produced by the cell, there was no corresponding shortage of hydrogen.

The simulations also indicate a close relationship between rising water saturation in the oxygen-side catalyst layer and GDL, and declining oxygen concentration. This increasing water saturation plays a key role in the mass transfer constraint that ultimate limits the maximum power and current produced by a PEM cell.

In the next subsection, the potential for changing input parameter settings in the ANSYS module to modify the current density at which the mass transfer constraint comes into play will be examined. The aim is to get a better fit between the ANSYS Module V-J curve for the large single PEM cell over the full range of current density, including the higher values, and the experimentally-measured performance.
6.3.3 Input parameters affecting mass transfer constraint

6.3.3.1 Input parameters identified

The analysis presented in the previous subsections has shown that oxygen concentration and water saturation play an important role in the mass transfer constraint in a PEM fuel cell. Both of these variables are outputs of the ANSYS Fuel Cell Module simulation. In the present subsection, various input parameters that can affect the onset of this constraint via oxygen concentration and water are investigated, namely:

- Cell temperature
- Operating pressure of reactant gas for both hydrogen and air
- Pore blockage for gas diffusion
- Pore blockage for transfer current
- Viscous resistance
- Porosity in the oxygen catalyst

6.3.3.2 Temperature and pressure effect on gas concentration and water saturation

The rate of diffusion of air through the oxygen-side GDL to the catalyst plays a critical role in determining the reaction rate on this electrode, and hence the resulting cell current. In the ANSYS Fuel Cell Module, gas phase species diffusivities can be computed by using the dilute approximation method. In this method, the following equation is used to compute the diffusivity of air:

\[ D_i = \varepsilon^{1.5} (1-s)^{r_s} D_i^0 \left( \frac{p_0}{p} \right)^{\gamma_p} \left( \frac{T}{T_0} \right)^{\gamma_t} \]  \hspace{1cm} (4.13)

where \( D_i^0 \) is the mass diffusivity of species of \( i \) at reference temperature and pressure \( (p_0, T_0) \). These reference values and the exponents \( (\gamma_p, \gamma_t) \) as well as the exponent of pore blockage \( (r_s) \) are defined in the Fuel Cell and Electrolysis user defined functions (UDF) as,

\[ p_0 = 101325 \text{ N/m}^2 \]
\[ T_0 = 300 \text{ K} \]
In this equation, it is clear that the gas diffusivity depends on pressure and temperature. Hence influence of changing the operating temperature and pressure of the cell on the current density at which the mass transfer constraint starts to have a major influence on the V-J curve will be explored.

Another way cell temperature and pressure can affect the onset of the mass transfer constraint is through their influence on the water condensation rate. A PEM fuel cell works at relatively low temperatures of less than 100 °C, so that some water vapour formed by the electrochemical reaction condenses to liquid form. Since water is generated on the oxygen side, at high current density the condensation to liquid water is greatest on this side. In ANSYS Module, the water saturation \( s \) - that is, the volume fraction of liquid water - is used to represent the water formation and transport. Liquid water plays the important role of hydrating the membrane, but it also blocks passages in the gas diffusion layer reducing the diffusion rate. Liquid water in the catalyst layer can reduce the effective reaction surface area on the catalyst. All these factors have a negative impact on overall cell performance.

In the ANSYS Module, the water condensation rate is calculated using the following equation (ANSYS, 2012):

\[
r_w = c_r \max \left( \left[ (1 - s) \frac{P_{wv} - P_{sat}}{R T} M_{H_2O}, [s \rho_l] \right] \right)
\]

where \( s \) is the water saturation, \( P_{wv} \) is the water vapour pressure, \( P_{sat} \) is the saturation pressure, \( \rho_l \) is the density of liquid water, the condensation rate constant is hardwired to \( c_r = 100 \text{ s}^{-1} \) (per second). This term is applied only inside the catalyst and gas diffusion layers.

As the equation 4.10 shows, the water condensation rate is dependent on temperature and pressure.

The simulation was therefore rerun under two different sets of conditions of temperature and pressure compared to the base case:
• Base case: temperature 300K, gauge pressure 1 bar
• Higher pressure case: temperature 300 K, gauge pressure 3 bar
• Lower temperature case: temperature 290 K, gauge pressure 1 bar

The resulting V-J curves are shown in Figures 6.17 (giving corresponding values for average oxygen concentration in the catalyst layer) and 6.18 (giving corresponding values for average water saturation).

![Figure 6.17: V-J curve for the base case, higher pressure gas case, lower temperature case, compared with the fitted experiment data. The average oxygen concentrations in the catalyst layer (kmol/m$^3$) at each point are also shown](image)

![Figure 6.18: V-J Curve for base case, higher-pressure gas, lower-temperature case, compared with the fitted experiment data with average and water saturation in the catalyst layer given at each point](image)
Figure 6.19: Water saturation versus current density curve for base case, higher-pressure case, and lower temperature case

Figure 6.20: Oxygen concentration versus current density curve for base case, higher-pressure case, and lower temperature case
For each simulated curve in Figure 6.17, the oxygen concentration dropped along the curve as current density increased, as shown in Figure 6.19. For each simulated curve in Figure 6.18, the water saturation increased along the curve as current density increased, as shown in Figure 6.20. For similar current density near 0.4 A/cm², oxygen concentrations of base case and lower-temperature case were very close, while oxygen concentration of higher-pressure gas was nearly three times of base case. At higher current density around 0.43 A/cm², the concentrations of higher-pressure case were also larger than two times of the value of base case. As a result, the oxygen concentration was related to the pressure a lot, but little to the temperature. The case was different for water saturation. For the similar current density 0.4 A/cm², the base case had the lowest water saturation. Both higher-pressure case and lower-temperature case had higher water saturation. As the equation 4.10 shows, the water condensation would increase with higher pressure and lower temperature.

Most notable, however, while changing the cell temperature and pressure did affect the shape of the V-J curve, at the higher current density end of the curves, the current density in all three cases still tended towards the same value, which was much higher than in the fitted experimental V-J curve (purple). Hence varying cell operating temperature and pressure in this way did not help in bringing the onset of the mass transfer constraint in the simulated V-J curves back towards the much earlier onset in the experimental curve.

### 6.3.3.3 Other factors

Other input parameters in ANSYS Module that may affect the limitation of the maximum current density through the mass transfer constraint including the following:

- The ‘pore blockage for gas diffusion’, corresponds to $r_s$ in equation 4.13. When $r_s$ is increasing, the effect of $s$ on diffusivity of air will be bigger. For the same $s$ value, the diffusivity of air will be smaller.

- The pore blockage for transfer current, $r$, which is used to account for liquid blockage to the reaction surface by modifying $R_{an}$ and $R_{cat}$ in equation 4.3 and 4.4, as follows:

$$R_{an}^{new} = (1 - s)^r R_{an}$$

$$R_{cat}^{new} = (1 - s)^r R_{cat}$$
Some with pore block for gas diffusion, while $r$ is increasing, the effect of $s$ on current densities will be bigger. For the same $s$ value, the current densities will be smaller.

- The viscous resistance, which was used to calculated pressure drop in porous media by ANSYS using Darcy’s law. Higher viscous resistance can cause higher pressure drop.

- The porosity in the oxygen catalyst, corresponds to $\varepsilon$ in equation 4.13. Higher porosity would lead to higher diffusivity of air.

The parameters are changed to reduce the current density, aiming at compress the whole curve to get lower current density limitation. The changed parameters are shown in Table 6.3.

<table>
<thead>
<tr>
<th>Name</th>
<th>Original value</th>
<th>Changed value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pore blockage for gas diffusion</td>
<td>2.5</td>
<td>10</td>
</tr>
<tr>
<td>Pore blockage for transfer current</td>
<td>2.5</td>
<td>10</td>
</tr>
<tr>
<td>Viscous resistance</td>
<td>1e+13</td>
<td>1e+20</td>
</tr>
<tr>
<td>Porosity in the oxygen catalyst</td>
<td>0.28</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Table 6.3: The values for other input parameters that can affect the mass transfer constraint used in the base-case simulation, and changed values explored in reruns of the simulation.

Figure 6.21: V-J curve for base case and other factor case, comparing with fitted experimental curve
The results of the new case were still not ideal. The current density at 0.5 V was reduced to 0.24 A/cm², but the current density at 0.4 V was raised to 0.45 A/cm². The V-J curve of other factor case crossed the curve of the base case. The change of these factors did reduce the average current density at 0.5 V to a lower value, 0.24 A/cm², which was on the extension of the experimental curve. But the increase in current density at 0.4 V showed that the experimental curve was still not being matched. Overall the changes to the four parameters taken together did not show any significant improvement in the representation of the mass transfer constraint on the V-J curve.

6.3.3.4 Inability to match mass transfer constraint in experimental V-J curve

From the investigations conducted in this section, it has been shown that the ANSYS Module can represent the turn-down of the V-J curve due to the mass transfer constraint in the end of V-J curve. The main reason of mass transfer constrain is considered as the increase in water saturation and decrease in oxygen concentration at high current densities. However, for the large single PEM cell studied here, it has not been found possible so far to change the values of input parameters to get an onset of this constraint at lower current densities, as was found in the experimental V-J curve.

Further changes in input parameters, probably changes of larger magnitude, still need to be explored to see if these can produce a simulated V-J curve with a mass transfer constraint occurring over a range of current density closer to what was found in the corresponding experimental curve. In addition, changes in other input parameters not so far investigated, most notably the parameter \( c_r \) in equation 4.10 that has such a key effect on the rate of water condensation, deserve to be studied. In the present ANSYS Module this parameter is fixed in the software and cannot be change.

6.4 Conclusion

A large PEM fuel cell has been simulated in the ANSYS Fuel Cell Module using the same input parameters as for the small cell C modelled in chapter 5, since the cell’s characteristics were the same, except that its active area was 45 times larger. The settings found for the best
fit curve for cell C from the simulation were used in the large fuel cell model. The channel configuration on the oxygen side, however, different to that in the small cell C.

The comparison of experimental and simulation results shows that the simulation represent the performance of large fuel cell in higher voltage part (Table 6.2) with small variances. But in voltage less than 0.6 V, the current density of simulation (0.4 A/cm$^2$ at 0.5 V) was much higher than the experimental data (0.24 A/cm$^2$ at 0.5 V). It suggested that the input parameters were suitable to represent the experimental value in the lower current density range, but the model did not consider power loss due to mass transfer limitation in the fuel cell in high current density.

As the current density figure shows, the maximum of current density is 0.4A/cm$^2$, and the area of maximum of current density was almost half of active area. For the current density distribution pattern and molar concentration pattern, it is clear that the lack of oxygen gas in the bottom lead to the lowest current density area in the membrane.

The mass transfer limitation, shown as the turn down of the V-J curve in the end of the curve, was studied. The mass transfer limitation showed in simulation result was happened in much higher current density than the experimental result. The Butler-Volmer equations used in the Module shows that the gas concentrations and water saturation are related to the mass transfer limitation. Temperature, pressure, and other input parameters were changed to find out the effect on the gas concentrations and water saturation, hence the current density. Since the relation of current limitation with other parameters have not been clearly found. As the graphs showed that the pressure and temperature would affect the water saturation and gas concentration, hence the current density. But the ANSYS simulation was not able to move the mass transfer limitation to the current density point when the curve of experiment begins to turn down.
7. SIMULATION MODELLING OF FUEL CELL STACKS IN THE ANSYS FUEL CELL MODULE

7.1 MODELLING FUEL CELL STACKS

Up to this point in the thesis, the focus has been entirely on single PEM fuel cells. In this chapter, the application of the ANSYS Fuel Cell Module to PEM fuel cell stack will be considered in a preliminary way. The fuel cell stack model is constructed in the module of a four-cell stack, where the individual cells have the same active area, catalyst materials, GDLs, and flow channel configuration as in the 5 cm² small cell C investigated both experimentally and through simulation in chapter 5. This model is then run to simulate the performance of the four-cell stack. The aim of the simulation is to investigate the effects of stacking single cells on the total system performance, and hence the average performance per cell. The latter can then be compared with the performance of the single cell C of the same size, design and materials.

The configuration of fuel cell stack is shown in the Figure 7.1. In ANSYS Fuel Cell Module, the bipolar plates are considered half as hydrogen endplate for one cell and half as oxygen endplate for the other cell. (The figure did not show the actual size of the fuel cell stack.)
For ANSYS Fuel Cell Module, fuel cell stacks were treated similar to single fuel cell. The difference was there would be one more setting option specially for fuel cell stack, shown in Figure 7.2. For the stack management, ANSYS Fuel Cell Module combined the endplates, channels, catalyst layers, gas diffusion layers, and membrane of each cell in the stack into a unit, named as ‘fcu-’ (shown in figure 7.1).

All the input parameters could be the same with the fitted value for cell C, expect the gas flow rate. When the current density was assumed the same with single fuel cell, the total gas consumption for the four-cell stack would be four times of the single fuel cell.
For the stack model, there were four through holes passing through all cells from one end plate to the other and supplying the flow channels in each cell. There were therefore only two gas inlets – one for hydrogen, and one for oxygen –. Similarly there were only two gas outlets in the stack model, one for each gas. The oxygen channels show in Figure 7.3. Similar configuration was applied to the hydrogen side channels, and the hydrogen supplied in the opposite direction of oxygen flow.

Figure 7.3: Oxygen channels for the stack model with gas flow direction

The ANSYS Fuel Cell Module simulation of even just a four-cell stack, with a relatively small active area per cell, was very intensive in terms of computing time and high-performance computer resources. Hence in the present study the simulation of the stack models had to be limited to a single voltage operating point, specifically 2.8 V (0.7 V per cell). At this point, the spatial distributions across the active area of each cell in the stack of current density, oxygen concentration, and hydrogen concentration, along with the gas
velocities in the channels on the oxygen and hydrogen sides in all cells, were generated and studied. The results obtained were then compared to the corresponding results for the 0.7 V operating point for the earlier single cell C.

In the future, clearly it will be important to extend the stack modelling to investigation of all variables over the full range of V-J operating points, and to obtain a full V-J curve. In addition, in the case of model it will be very relevant to compare the performance of all the individual cells to see if there is significant variation depending on their location in the stack.

It would also be interesting to compare a fuel cell stack of a given total active area with a single fuel cell of the same active area, as well as stacks with the same total active area but different numbers of cells.

7.2 MODEL OF PEM FUEL CELL STACK

7.2.1 Model setup

The four-cell stack had one hole passing through all the cells to supply the hydrogen-side inlets, and a second through hole connected to all the hydrogen-side outlets of the cells. A second pair of through holes served the same purposes for the air supply and outlets (as shown in Figure 7.3). Each through hole had one end open and the other end closed. Hence the hydrogen gas was supplied just from one end and the flow divided along the through hole flowing into the hydrogen-side channel configuration of each cell. The same situation applied to the oxygen supply. Hence in this model it was possible that gases could be fed a different flow rate to each cell. All four cells in stack were connected in series, so that the current through each cell must be the same. The voltage for each cell was again assumed to be equal (0.7 V) giving a stack voltage of 2.8 V.

The geometry and meshing used in the model of fuel cell stack is shown in Figures 7.4 to 7.6.
Figure 7.4: Geometry in the model of the four-cell stack

Figure 7.5: Overall view of the mesh in the models of stack.
The input parameters or the cell characteristics were also kept the same as for single fuel cell C (Table 7.1).

<table>
<thead>
<tr>
<th>Anode</th>
<th>Unit</th>
<th>Cathode</th>
<th>Unit</th>
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</thead>
<tbody>
<tr>
<td>Ref. Current Density</td>
<td>100 A/m²</td>
<td>Ref. Current Density</td>
<td>10 A/m²</td>
</tr>
<tr>
<td>Ref. Concentration</td>
<td>0.04 kmol/m³</td>
<td>Ref. Concentration</td>
<td>0.03 kmol/m³</td>
</tr>
<tr>
<td>Concentration exponent</td>
<td>1</td>
<td>Concentration exponent</td>
<td>1</td>
</tr>
<tr>
<td>Exchange Coefficient (a)</td>
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<td>Exchange Coefficient (a)</td>
<td>0.1</td>
</tr>
<tr>
<td>Exchange Coefficient (C)</td>
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<td>1.9</td>
</tr>
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<td>Porosity(GDL)</td>
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<td>Viscous Resistance(GDL)</td>
<td>1E+12 1/m²</td>
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<td>Porosity(CL)</td>
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<td>Porosity(CL)</td>
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<td>viscous Resistance(CL)</td>
<td>1E+12 1/m²</td>
<td>viscous Resistance(CL)</td>
<td>1E+12 1/m²</td>
</tr>
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<td>surface/Volume Ratio</td>
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<td>20000 1/m</td>
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<td>Oxygen inlet</td>
<td>7e-6 kg/s</td>
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<table>
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<th>Membrane</th>
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<th>Unit</th>
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</thead>
<tbody>
<tr>
<td>Equivalent Weight</td>
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<td>Open-Circuit Voltage</td>
<td>1 V</td>
</tr>
<tr>
<td>Protonic Conduction Coefficient</td>
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<td>Specific Leakage Current</td>
<td>0 A/m³</td>
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<tr>
<td>Protonic Conduction Exponent</td>
<td>0.5</td>
<td>Electrolyte Project Area</td>
<td>0.000506 m²</td>
</tr>
</tbody>
</table>

Table 7.1: Input parameters for the simulation of four-cell stack
7.2.2 Outputs and Analysis

For a stack voltage of 2.8 V, the final total current of the stack was 0.44 A, the average current density of 0.087 A/cm². While the integrated current across the active area of each cell was the same, the distribution of current density was found to vary from cell to cell (as shown as in Figures 7.7). The current density distributions in the membranes of the two outer cells were different to those of the middle two cells. The end cells had a higher current density in the outer part of the active area, and lower current density in the middle. The middle two cells had higher current density in the middle and lower current density in the outer region, exclude the vertical edge.

Figure 7.7: Maps of the distribution of current density in each cell membrane in stack

From the Figure 7.7, the layers in the end had a total different distribution with the layers in the middle. The two layers in the end had higher current density outside and lower current density in the middle. The middle two layers had higher current density in the middle and lower current density outside.
As Figure 7.7 shows, the current density distribution in the membrane of each cell was different, with point made earlier that the two end cells had broadly opposite distribution patterns to the two cells in the middle. The maximum current density found along the first and last channel in cell, was around 0.1 A/cm², 15% larger than the average current density for the stack as a whole. The minimum current density was about 0.08 A/cm², at the middle of in cell 1 and cell 4, corners in cell 2 and cell 4, 7% lower than the average current density. Compare to the current density distribution of cell C of 0.7 V (Figure 5.29 c). Although the average current density was lower than the cell C. The highest current densities were both at

Figure 7.8: Current density distributions in the membrane of each cell
two side (vertical) of membrane active area. The current densities of two endplates had the same trends with the cell C, higher current density outside and lower current density inside.

The hydrogen concentration distributions in the hydrogen-side catalyst layer differed from cell to cell, as did the average hydrogen concentration across the full active area of the cell, as shown in Figures 7.9 and 7.10. The first cell’s catalyst layer had an average hydrogen concentration of 2.24e-2 kmol/m³ while the fourth layer had an average hydrogen concentration a little larger at 2.46e-2 kmol/m³. The average hydrogen concentration in the catalyst layers of the middle two cells was significantly lower at 1.61e-2 kmol/m³. In each layer, the highest hydrogen concentration was in the middle of the active area, and the lowest towards the four corners.

Figure 7.9: Hydrogen concentration in the hydrogen catalyst layers of each cell
Oxygen concentrations in the oxygen-side catalyst layer also differed from cell to cell (Figures 7.11 and 7.12). The catalyst layers in the two middle cells middle had lower oxygen concentrations than the plates at two ends. The average oxygen concentrations in the catalyst layers of the two cells in the middle of the stack (cells 2 and 3) were around 30% lower than the oxygen concentrations of the two end cells. For all cells, the oxygen concentration was higher in the top and lower in the bottom of the active area, since the air was fed in from the top. The differences between average value with maximum value and minimum value were about 2e+2 kmol/m³ for all cells, and less 10% of the average oxygen concentration.
Figure 7.1: Oxygen concentrations in the oxygen-side catalyst layers of each cell in stack

(a): cell 1  
(b): cell 2
Figures 7.13 and 7.14 show the gas velocity respectively in the oxygen-side and hydrogen-side channels of each cell, including through holes. The gas velocities in the channel for each cell were not the same in the stack model. Both for hydrogen side and oxygen side, the cells farthest from the gas inlet had the highest gas velocity, and those gas channels in the middle had lower gas velocity. The third cell from the gas inlet had the lowest average gas velocity in the case of both hydrogen and oxygen.

Figure 7.13: Gas velocity in oxygen-side channels: the overall through-hole and channels in each cell (left); and just in the oxygen-side channels of each cell (right)
7.3 Conclusions

In this chapter, the ANSYS Fuel Cell Module has been applied in a preliminary way to modelling of a four-cell PEM fuel cell stack. The stack model had a single through hole for supplying hydrogen in turn to the hydrogen-side channels of each cell from one end, and another similar through hole for air supply. Similarly through holes were used as the outlets from the hydrogen- and air-side channels for each cell respectively.

The simulation of fuel cell stack is aiming to compare the current density of fuel cell stack and single fuel cell (Cell C) and study the difference of different layer of a fuel cell stack. The fuel cell stack model showed poorer performance than the single fuel cell model, the average current density of the stack model was less than half of the average current density of sing fuel cell model.

The result of the stack model shows barely opposite current density distribution between cells in the two ends and cells in the middle. The current density distribution was related to the gas distribution. Different from the single fuel cell mode, the result of simulation of the stack model shows the significant difference of gas distribution between cells. The middle plates had lower gas concentration than the two endplates. The effect of gas concentration might lead to the different current density distribution. The figures of gas concentration and gas velocity showed that the gas had not been send to every layer of the fuel cell stack evenly. That is the reason why the current density distribution of the two middle cells was quite different from single cell C. The channels need to be redesigned to gain more even gas distribution.

In the ANSYS Fuel Cell Module, the fuel cell stack models assumed that each cell had the same electrical potential difference. This appears to be a built-in assumption for stacks in the version of the Module used in the present study. Variations in the current density distribution were, however, found between the four cell stack. Moreover, experiments conducted at RMIT on PEM fuel cell stacks have shown that there are usually some differences in the voltages of
the individual cells. Hence the a priori assumption the ANSYS module applied to stacks of equal voltage for each cell would appear to be an unrealistic restriction, which needs to be relaxed in future versions of the Module.

Because the ANSYS Module simulation of multiple-cell stacks requires so much computing time on high-performance computers resources, the stack modelling in the present chapter has had to be limited to a single voltage operating point, In the future, clearly it will be important to extend the stack modelling to investigation of all variables over the full range of V-J operating points, and to obtain a full V-J curve. In addition, in the case of model B it will be very relevant to compare the performance of all the individual cells to see if there is significant variation depending on their location in the stack.

It would also be interesting to compare a fuel cell stack of a given total active area with a single fuel cell of the same active area, as well as stacks with the same total active area but different numbers of cells.
8. CONCLUSION AND RECOMMENDATIONS

8.1 INTRODUCTION

In this chapter, answers are provided (in section 8.2) to the research questions set for this thesis in chapter 1, based on all the research conducted and reported in the earlier chapters. Additional conclusions drawn from this work are presented in section 8.3, and recommendations made for future research and applications within the general area of computer-simulation modelling of fuel cells in section 8.4.

8.2 RESPONSE TO THE RESEARCH QUESTIONS

8.2.1 To what level of accuracy can simulations of the performance of individual PEM fuel cells using the ANSYS Fuel Cell Module match experimental performance data?

In this thesis, the ANSYS Fuel cell Module has been used to simulate small single fuel cells (active area 5 cm²), and a large single fuel cell (225 cm²), and the VI curves obtained as an output from the simulation have been compared to experimental VI curves for cells with the same geometry and material properties, and corresponding active areas. No experimental data have been obtained covering other variables such as gas concentrations and velocities within the channels of the cells, and water saturation.

The simulation of small single fuel cell A (area 5 cm²) was compared with prior experimental result done by Doddathimmaiah (2008). The input parameter settings for the simulation were taken from reference current densities and exchange coefficients in a mathematical model fitted to the experimental result by Doddathimmaiah and Andrews (2008). The V-J curve
obtained from the ANSYS simulation showed similar trend and current density range to the earlier experimental curve. The start point at 0 A/cm² was 0.75 V for the experiment, and 0.8 V in the ANSYS simulation, that is, 7% higher. The end point at 0.12 A/cm² was 0.1 V for the experiment, and very nearly the same for the ANSYS simulation. The maximum difference in the voltage between simulation and experiment was about 0.09 V at 0.07 A/cm², the simulation result being 30% larger than the experimental one. Current density of cell A was never high enough for the mass transfer constraint to play a role.

The simulation of cell B used the same geometry as cell A. Since the material of membrane, catalysts and gas diffusion layers were different, reference current densities and exchange coefficients were changed to get the best fit of the simulation result to the experimental result. After the fitting procedure, the V-J curves of the simulation and experiment results were generally very close. Although the experimental result had a higher near-open circuit voltage: when the current density was very small at 0.0014 A/cm², the experimental voltage was 0.864 V while the corresponding voltage in the simulation was 0.862 V. The end points of the curves were at the same point 0.098 A/cm² at 0.6 V. Between 0.02 and 0.09 A/cm², the voltage of the ANSYS simulation result was only about 3% lower than the experiment result.

The simulation of cell C was done by reducing the width of the gas channels for both experiment and simulation. The simulation result was again modified by changing input parameters to get the best fit to the experimental result. The fitted V-J curve was very close to the experimental curve from 0 A/cm² to 0.28 A/cm². Then the voltage of the simulation was higher than in the experiment to the end of the curve. The maximum current density of the experiment was 0.37 A/cm² at 0.4 V, while the simulation voltage at 0.37 A/cm² was about 0.5 V, 25% higher than the experimental voltage. In the experimental V-J curve of cell C, there was a clear sign of the turndown in voltage at higher current density, indicating the onset of mass transfer constraint. However, in the simulation of cell C, water saturation was not considered at this stage, and this explains why the simulation did not show any signs of a mass transfer constraint and had a voltage much higher than in the experimental V-J curve at high current density.

The simulation of the large fuel cell (225 cm² active area) used the fitted input parameters of cell C since the large cell had the same material in this membrane electrode assembly and GDLs. The smoothed experimental V-J curve and the simulation curve were almost the same from 0.06 to 0.14 A/cm², while below 0.06 A/cm², the voltage on the simulation curve was a
little higher (less than 5%) than the experimental curve. When current density was higher than 0.14 A/cm², the smoothed experimental curve showed evidence of the mass transfer constraint by turning down more rapidly, but the simulation curve still continued a straight linear decline. At the maximum current density of on the experiment curve of 0.24 A/cm², the voltage was 0.51 V, while the voltage of the simulation was 0.64 V, 25% higher than the experimental result.

In summary, ANSYS Fuel Cell Module was able to simulate the single PEM fuel cells with the right input parameters, with a relatively small difference of only a few percent between experimental and simulated voltages at the same current density over most of the current density range. But at higher current density when the mass transfer constraint began to play a role, the difference between simulated and experimental curves became increasingly large.

8.2.2 How does changing the membrane active area affect the performance of a single PEM fuel cell in terms of current density and power density?

In this thesis, a large single fuel cell (225 cm² active membrane area) was modelled and experimental measured to compare with the small single fuel cell C (5 cm² active area). The large cell used the same materials of in the membrane, catalysts and gas diffusion layers as the small cell C. The input parameters for the simulation of the large fuel cell were the same as those for the simulation of cell C, except for the inlet gas flow rates.
The experimental V-J curve showed that the current density of large single fuel cell was always lower than the small single fuel cell C, with a maximum drop of 19.5% for the small fuel cell at 0.24 A/cm$^2$ (Figure 8.1). The maximum power density of large single fuel cell was 0.123 W/cm$^2$, while the maximum power density of small single fuel cell C was 0.166 W/cm$^2$. The power output of large single fuel cell was 26% lower than the small single fuel cell C.
A comparison of the simulated V-J curves for the small and large fuel cell is shown in Figure 8.2. From the beginning of the curves, the voltage of the large single fuel cell is lower than the voltage of small single fuel cell C at a given current density. The maximum voltage difference is about 0.05 V at 0.14 A/cm$^2$, and the associated power density difference of 0.007 W/cm$^2$. In the Figure 8.2, the voltage on the simulated V-J curve for the large cell and small cell had dotted line in the end part as unfitted part, because the simulation results were much higher than the experimental result.

Comparing the current density distribution of large single fuel cell and small single fuel cell at 0.7V in Figure 8.3.
The current density of small single fuel cell is 5% variation of average current density (0.192 A/cm$^2$); the large fuel cell is 50% variation of average current density (0.137 A/cm$^2$). The maximum current density was 0.202 A/cm$^2$ for the small fuel cell, 0.211 A/cm$^2$ for the large fuel cell. As a result, the average current density of large fuel cell was lower than the small fuel cell.

From the comparison of V-J curves of experiment and simulation for small single fuel cell (5 cm$^2$) and large single fuel cell (225 cm$^2$), the drop in voltage of large fuel cell was found for same current density along the V-J curves. As a result, it could be answer to the research question that, with increasing of the membrane active area, the voltage would drop for the same current density, hence the overall power output.

### 8.2.3 How does the performance of a PEM fuel cell stack change compared to single fuel cells in terms of current density and power density?

A four-cell PEM fuel cell stack was modelled using ANSYS Fuel Cell Module. Each cell had the same design as the small fuel cell C and a 5 cm$^2$ active area. The channels of the same side of each cell were connected by through holes to their respective inlets and outlets on the end plates, in the normal manner in multi-cell stacks. Hence the stack had one inlet and one inlet on each endplate. The gas flow rates for hydrogen and oxygen were about four times corresponding values used when modelling the small single fuel cell C. The comparison of
the fuel cell stack to the single fuel cell C was done at 2.8 V, and included analysis of the performance of each cell of the stack.

In the simulation, the total current of the fuel cell stack at 2.8 V was 0.44 A, and the average current density of all cells was 0.087 A/cm². By contrast, the average current density of the small single fuel cell was 0.18 A/cm², over twice that for the fuel cell stack.

Inspection of the maps of current density across each cell in the stack showed that:

- the variation for cell 1 was from 0.08 A/cm² to 0.101 A/cm²;
- for cell 2, form 0.076 A/cm² to 0.098 A/cm²;
- for cell 3, from 0.078 A/cm² to 0.098 A/cm²; and
- for cell 4, from 0.082 A/cm² to 0.105 A/cm².

In the simulation of small fuel cell C at 0.7 V, the current density varied between 0.188 A/cm² to 0.202 A/cm², that is, between much higher values than for the cells in the stack. Comparing the current density distributions of stack cells and the single cell, the small fuel cell had higher current density towards the outside of the active membrane area, being highest at the top corners. The current density in the fuel cell stack had two kinds of distribution. In the two end cells, the current density was higher on its two sides and lower in the middle, and then dropped steeply towards the centre; while for the two cells in the middle, the highest current density was also at the sides, while decreasing from the centre to edge.

Clearly, the average current density in the fuel cell stack was much lower than in the single fuel cell. A possible reason may have been that the average oxygen concentration in the catalyst layer of the fuel cell stack was much lower than the average oxygen concentration in the small single fuel cell C (Figure 8.4).
Figure 8.4: Oxygen concentration in the catalyst layer of cell C (left) and in the individual cells in the four-cell stack (right)

Although the gas flow rates to the four-cell stack were four times larger than to the single cell, the distribution to each cell in the stack was not even. More gas was fed into the cell 1 and cell 4 in the stack, compared to the middle two. It can be seen from Figure 8.4 that the oxygen concentration in the catalyst layer of cell C ranged from 1.10e-02 to 1.31e-02 kmol/m³, while in the four-cell stack the oxygen concentration varied from 3.56e-03 to 4.03e-03 kmol/m³ in cell 1; from 2.24e-03 to 2.74e-03 kmol/m³ in cell 2; from 2.47e-03 to 2.84e-03 kmol/m³ in cell 3; and from 3.56e-03 to 4.03e-03 kmol/m³ in cell 4. Hence generally the oxygen concentrations in the catalyst layers of the four-cell stack were much lower than those in cell C. The hydrogen concentrations were similarly lower in the stack cells. These lower concentrations are likely therefore to have caused a lower reaction rate, and hence lower current density in the stack. In particular, the lower oxygen concentration in the middle cells in the stack would have limited overall performance of the stack, since the stack current would be limited by the current through the worst performing cell. It is also noteworthy that since the voltage of each cell in the stack was set equal by the ANSYS Fuel Cell Module the drop in current due to a low oxygen concentration would be greater than if this voltage had been allowed to fall.

In summary, according to these simulations, the average current density in a four-cell stack, was only around half that in a single cell of exactly the same design. The likely reason is that
the uneven gas distribution to the four cells in the stack led to particularly low oxygen concentration in the middle cells. The resulting lowered current density would then have limited the total current through the whole stack.

It was also noted that in the ANSYS Fuel Cell Module simulation of the stack, the voltage of each cell in the fuel cell stack was set to be the same, independently of the current density and gas flow distributions. However, actual experimental measurements show that the voltage of each cell in a stack can vary. Hence this constraint in the ANSYS Module needs to be relaxed, so that the voltage of each cell in a stack is an output of the overall simulation rather than a fixed input assumption.

8.2.4 What is the best approach to using the ANSYS Fuel Cell Module to represent the mass-transfer limited portion of the voltage current density curve of a PEM fuel cell at higher current densities?

From the investigations conducted in chapter 6.3, it has been shown that the ANSYS Module can represent the turn-down of the V-J curve due to the mass transfer constraint towards the end of a V-J curve. The main cause of the mass transfer constrain is identified as the increase in water saturation in the oxygen-side gas diffusion layer, and consequent decrease in oxygen concentration at high current densities. However, for the large single PEM cell studied using the ANSYS Module here, it has not been found possible so far to change the values of input parameters to get an onset of this constraint at lower current densities, as was found in the corresponding experimental V-J curve.

Further changes in input parameters, probably changes of larger magnitude, still need to be explored to see if these can produce a simulated V-J curve with a mass transfer constraint occurring over a range of current density closer to what was found in the corresponding experimental curve. In addition, changes in other input parameters not so far investigated, most notably the parameter $c_r$ in equation 4.10 that has such a key effect on the rate of water condensation, deserve to be studied. In the present ANSYS Module this parameter is fixed in the software and cannot be change.
8.3 CONCLUSIONS

Additional conclusions drawn from the research conducted for this thesis, over and above those covered in the previous section as answers to the research question, are presented in this section.

In the ANSYS Fuel Cell Module, it was found that the same values for the transfer coefficients ($\alpha_{\text{cat}}$ and $\alpha_{\text{an}}$) were used in both the oxygen-side and hydrogen-side equations. However, it was pointed out in this thesis that the exchange coefficients used respectively on the two electrodes of a cell should be different. Hence it was concluded that a modification to the script describing the Butler-Volmer equations in the ANSYS Module software needed to be made to introduce new variables to allow separate and independent setting of the two exchange coefficients for each electrode. This modification was done in ANSYS 14.5 and the results obtained were successful.

By changing the reference current density and exchange coefficient, the ANSYS Fuel Cell Module correctly showed the effect of these parameters on the performance of a variety of cells. It was found that increasing the reference current density raised the V-J curve, and that all the V-J curves for the different reference current densities were approximately parallel with each other. As the exchange coefficient was decreased, the V-J curve was lowered, and the slope downward of the curve was enhanced.

Changing the flow channel configuration for the small single fuel cell to narrower channels was found to increase the current output. The current density distribution of small single fuel cell A showed the significant current density gap between channels and in the middle of the channels. Lowering width of the channels and ribs in small cell C was found to increase the total performance compared to cell A.

From the simulations done with the ANSYS Fuel Cell Module in this thesis, some limitations were found:

- The onset of the mass transfer constraint in simulated V-J curves generally occurred at much higher current densities than found in the experimental results for the same cell. It was not found possible in the present work to change the input parameters to the Module to move the mass transfer constraint to the right place. Some input
parameters that may affect the mass transfer constraint, such as $c_r$, were not able to be changed in the present version of the Module.

- The voltages of all cells in a stack were assumed equal in the fuel cell stack model in the ANSYS Fuel Cell Module, at the value of the total stack voltage fed in as an input to the Module divided by the number of cells in the stack. However, in actual experiments on fuel cell stacks, it was found that the voltages of each cell were not necessarily the same, with some dropping to near zero while others were at 0.6 V under extreme conditions. The automatic setting of equal cell voltages in a stack thus needs to be changed in the Module.

8.4 RECOMMENDATIONS

The following recommendations are made relating to the further development of simulation of PEM fuel cell.

More research needs to be done on the relation of flow rate with the gas concentration, and hence the current density in single PEM fuel cells, and for each cell within a stack.

Further study is also needed on the influence on PEM cell performance of flow channel width and distance between channels by varying these dimensions while keeping the gas concentration constant.

There needs to be more research conducted into what input parameters can be changed, or actual changes made to the basic code, in order for the ANSYS FC Module to represent more accurately the onset and magnitude of the mass transfer constraint and its relation to water saturation in the VI curves for single PEM fuel cells.

Further systematic research using ANSYS Fuel cell module simulations should be conducted into evaluating the relative effects on performance of increasing the active membrane area per cell, versus increasing the number of cells of a given area in a stack, while keeping the increase in total active area constant, with validation of the findings against experiments.

Further experience should be gained in the use of computer simulation modelling of PEM fuel cell stacks, by the ANSYS Module or other software, to optimise design features such as:

- Flow channel configuration on both hydrogen and oxygen sides
- Oxygen and hydrogen GDL material and geometric properties
- Catalyst properties such as specific surface area (that is, active area per unit volume)
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198


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Conference paper

Title: Experimental and numerical investigation of the impact of gas channel configuration on the performance of a small scale PEM fuel cell

Murphy K.K. Lai, Xiang Wu, Sherman C.P. Cheung, John Andrews

20th Australasian Fluid Mechanics Conference, Perth, Australia, 5-8 December 2016

Journal papers (planned)

1. Computer simulation modelling and experimental investigation of single PEM fuel cells with various membrane active areas

Xiang Wu, John Andrews, Bahman Shabani, Sherman C.P. Cheung

To be submitted to International of Hydrogen Energy

2. The influence of gas flow on PEM fuel cell stack performance: A computer simulation investigation using the ANYS Fuel Cell Module

Xiang Wu, John Andrews, Bahman Shabani, Sherman C.P. Cheung

To be submitted to Journal of Power Sources