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# Investigation of species transport in a gas diffusion layer of a polymer electrolyte membrane fuel cell through two-phase modelling

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#### Abstract

A two-phase polymer electrolyte membrane fuel cell model has been developed to investigate transport of species in a gas diffusion layer taking into account effects of liquid water saturation. A set of governing equations for mass, momentum, species concentration involving oxygen, hydrogen, water vapour and liquid water together with electrochemical reaction equations have been solved under computational fluid dynamics technique. The effects of presence of liquid water on the effective diffusivity of species have been investigated. A thorough comparison study of liquid water saturation model using power law with various exponential factors and a percolation based model has been carried out. The simulation results show that the power law model with exponential factor of 2 provides a good representation of species diffusivity and produces much closer agreement with experimental cell voltage, while the percolation based model produces overprediction of cell voltage. The effects of isotropic and anisotropic permeability of gas diffusion layer have also been studied and the simulated results show that the high isotropic permeability or a combination of high in-plane and low through-plane permeability results in higher performance of a polymer electrolyte membrane fuel cell. The fuel cell performance significantly deteriorates with low in-plane and high through-plane permeability of gas diffusion layer.

**Keywords:** PEM fuel cell; saturation; diffusivity; permeability; anisotropy; gas diffusion layer.

#### 1.0 Introduction

One of the major challenges facing the scientific and engineering communities is finding sustainable and environmentally friendly energy sources. Current energy demands are primarily met by burning fossil fuels. Over reliance on fossil fuels not only increases greenhouse gas emission, but also leads to depletion of the source. In search of alternative and clean energy source, much research has been focused on wind turbine, tidal energy, biomass and hydrogen fuel cells. Fuel cells have many advantages over the other sources such as continuous supply of energy without depending on nature, having compact design and wide ranges of applications. Among many different types of fuel cells, polymer electrolyte membrane (PEM) fuel cell

seems to be the front runner for deployment in variety of applications such as residential combined heat and power, automobiles, standby power unit and power plant.

A PEM fuel cell works by combining hydrogen and oxygen through electrochemical reaction and producing water. A fuel cell is made of the following components: flow channels, gas diffusion layers (GDL), catalyst layers and a membrane. A fuel cell is split into cathode and anode sides. Reactants flow through the flow channel, diffuse through the GDL and reach the catalyst layer. At anode side, hydrogen splits into proton and electron; proton is carried through the membrane to the cathode side, while electron flows through the outer circuit providing power and then reaches the cathode catalyst layer. At cathode catalyst layer, proton, electron and oxygen combine to produce water. Though the process seems simple enough, there are many issues that are preventing wider deployment of PEM fuel cell. One of these issues is high cost of PEM fuel cell due to the use of expensive platinum catalyst. One method for reducing the high cost would be to obtain increased performance, especially at higher load (higher current densities) from the existing architecture. In order for this to happen a thorough understanding of transport of species through flow channels, gas diffusion layers, electrochemical reactions at catalyst layers, charge transports, heat transport and phase change of water is required. Due to confined and highly reactive nature of fuel cell, experimental data of common parameters such as temperature, species distribution, pressure and saturation within a fuel cell is sadly lacking [1]. In this respect a computational fluid dynamics model can provide a great deal of insight into the processes and phenomena inside a PEM fuel. Since the working of a PEM fuel cell involves many complex processes, the accuracy of a computational fluid dynamics fuel cell model would depend on the accuracy of all the physical submodels.

The earliest pioneering models developed by Bernardi [2], Bernardi and Verbrugge [3], Springer et al. [4] and Nguyen and White [5] were semi-empirical, often onedimensional and mainly based on experimental data for membrane for calibrating the Further development in modeling studies led to performance characteristics. development of computational fluid dynamics based techniques. However, in the early days of developing computational fluid dynamics model for PEM fuel cells, singlephase models were developed with water was assumed to be present in the vapour phase. Mazumder and Cole [6], Nguyen et al [7] and Sivertsen and Djilali [8] have reported a three-dimensional single phase CFD modelling study of a PEM fuel cell. Lum and McGuirk [9] have investigated the effects of electrode thickness, shoulder width, degree of permeability of gas diffusion layer and oxidant concentration on the cell performance. Berning and Djilali [10] have investigated the effects of operational parameters such as temperature, pressure, stoichiometric flow ratio as well as geometric and material parameters such as electrode thickness, porosity and the channel width and land area. In these studies, water was considered to be present only in the vapour form. Though the studies provided excellent qualitative information, the real situation of the presence of liquid water inside gas diffusion layers, flow channels and its effect on the gas transport has not been studied. In the work of Wang et al [11], and You and Liu [12] both liquid and vapour phase of water flows have been considered, but only inside the cathode gas diffusion layer. In a follow-up paper, You and Liu [13] reported a two-dimensional, two-phase coupled PEM model, which showed that the liquid water also influenced the oxygen transport. Natarajan and Nguyen [14] employed a pseudo three-dimensional model to investigate liquid water movement inside cathode electrode. Berning and Djilali [15] also developed a three-dimensional, multi-phase, multi-component model considering heat and mass transfer. In their study liquid water transport inside the GDL was

numerically modeled by using viscous and capillary effects. This method was also implemented by Mazumder and Cole [16]. Min [17] developed a three dimensional, two-phase, non-isothermal model based on two-fluid model. Meng [18] developed a mixed domain two-fluid PEM fuel cell model, where water transport through the membrane was calculated by solving a conservation equation for water content in the membrane. With the advancement of knowledge and computer power, the recent trend has been to employ three dimensional, two-phase modeling. Khan et al. [1] have provided a recent review on two-phase flow modelling of PEM fuel cell and highlighted that anisotropic GDL properties, water saturation and inter-phase change between humidified gaseous species and liquid water are still less understood.

Despite having major advancement in developing computational model for PEM fuel cell, a complete fuel cell model is still illusive, because of the complexity of problem and further study is clearly needed in many aspects of sub-model development. The present study focuses on the investigation on the accurate modelling of the effective diffusivity of gas diffusion layer. The main function of a gas diffusion layer of a PEM fuel cell is to diffuse reactants towards the catalyst layer and remove product from the catalyst layer. Since a gas diffusion layer is made of randomly distributed fibres, it exhibits anisotropic behavior due to the orientation of fibres [19]. However, common trend has been to use isotropic Bruggeman correlation for treating the effective diffusivity of species for porosity effects. Several other diffusivity models have also been proposed [20-23] and a comprehensive comparison study by Hossain et al [24] using a single phase model shows that a percolation based anisotropic model where in-plane diffusivity is higher than through plane diffusivity provides a much closer prediction to the experimental performance curve.

During operation of a PEM fuel cell at high current densities, condensed water in the gas diffusion layer blocks pore spaces and reduces the effective gas diffusivity. The effect of saturation on gas diffusivity has been tackled by researchers through applying a power law relative diffusivity model. However, there are many differences in the value of exponent used in the power law model. For example, He et al [25] and Mishra and Wu [26] used a value of 3, whereas Jung et al [27] used a value of 1.5. Nam and Kaviany [28] have provided a detailed analysis of the effective gas diffusivity using a pore network model and shown that a combination of percolation based model for porosity effect and a power law with an exponent of 2 for saturation has provided a good agreement with the results obtained from the pore network model. Dawes et al [22] have investigated the effective diffusivity effects and reported that the percolation based models for both porosity and saturation have provided a better agreement compared to the power law model with exponent of 1.5.

The main aim of the present study is tackle the uncertainty involved in the relative diffusivity modeling. A comparative study of various power law and percolation models for saturation as well as the effects of isotropic and anisotropic permeability of gas diffusion layer has been reported here. A detailed description of the development of a two phase computation fuel cell model has also been provided. The present study highlights the importance of accurate description of sub-models under the overall computational fuel cell model.

#### 2.0 Mathematical Model

The governing equations for the PEM fuel cell model consist of continuity, momentum and species transport inside the catalyst layers, gas diffusion layers and the flow

channels. To represent the electrochemistry and transport phenomena through the membrane, appropriate source terms are applied at the anode and cathode catalyst layers. The set of governing equation in the present fuel cell model development is given below:

2.1. The mass conservation equation (continuity equation):

$$\nabla \left( \rho \vec{u} \right) = 0 \tag{1}$$

where  $\rho$  is the fluid density,  $\vec{u}$  is the velocity vector and  $S_m$  is the source term.

2.2 The momentum conservation equation:

$$\nabla \left( \rho \vec{u} \vec{u} \right) = -\nabla P + \nabla \left( \mu \nabla \vec{u} \right) + S_{u} \tag{2}$$

where *P* is the pressure and  $S_u$  is the source term.

In the flow channel,  $S_u$  is zero. In the gas diffusion layers and the catalyst layers Darcy's law term is added to the momentum equations to represent the momentum related to the porous media. This source term is expressed as:

$$S_u = -\frac{\mu \vec{u}}{\kappa} \tag{3}$$

where, K is permeability of the porous media (gas diffusion layers and catalyst layers).

2.3 The species conservation equation:

$$\nabla(\rho \,\vec{u} \,X_k) = \nabla \left( D_k^{eff} \rho \nabla X_k \right) + S_k \tag{4}$$

where index *k* refers to oxygen, hydrogen, water vapour and liquid water.  $X_k$  is the molar concentration of species *k* and  $D_k^{eff}$  is the effective diffusion coefficient of species *k*.

The source terms ( $S_k$ ) in the species conservation equation (4) are defined as zero for all regions of the model except at the catalyst layers. Species source term for anode and cathode catalyst layers are expressed as:

Consumption of hydrogen due to electrochemical effects at the anode catalyst layer

$$S_{H_2} = -\frac{IA}{2F}M_{H_2} \tag{5}$$

Consumption of oxygen due to electrochemical effects at the cathode catalyst layer

$$S_{O_2} = -\frac{IA}{4F} M_{O_2} \tag{6}$$

Production of water and flux of water due to electrochemical effects at the cathode catalyst layer

$$S_{cw} = \frac{[1+2\alpha]IA}{2F} M_{H_2O}$$
(7)

The flux of water due to electrochemical effects at the anode catalyst layer

$$S_{aw} = -\frac{\alpha IA}{F} M_{H_2O} \tag{8}$$

The current density *I* and net water transfer coefficient  $\alpha$  are used to determine these source terms.

To account for phase changes between water vapour and liquid water, the following source term has been added in the species transport equation for liquid water and water vapour (4) according to [29]

$$S_{wlp} = -S_{wvp} = \frac{(P_{wv} - P_{wv}^{Sat})}{RT} \times M_{H_2O} \times k_c$$
(9)

Where  $k_c$  is the water vapour condensation rate.

The diffusion coefficient of species can be expressed as [30]:

$$D_k = D_{k,ref} \left(\frac{T}{T_{ref}}\right)^{3/2} \left(\frac{P_{ref}}{P}\right)$$
(10)

where  $D_{k,ref}$  is the reference value at  $T_{ref}$  and  $P_{ref}$ . The diffusivity values obtained from equation (10) needs to be corrected for porous regions.

The diffusion coefficient in the porous regions can be expressed as

$$D_k^{eff} = f(\epsilon)g(s)D_k \tag{11}$$

Tomadakis and Sotirchos [20] model has been shown to provide the best representation of effective diffusivity and this percolation theory based diffusion model for random fibrous porous medium is given by [28]

$$f(\varepsilon) = \varepsilon \left(\frac{\varepsilon - \varepsilon_p}{1 - \varepsilon_p}\right)^{\alpha}$$
(12)

where  $\varepsilon_p$  is the percolation threshold and equal to 0.11.  $\alpha$  is the empirical constant which depends on the direction.  $\alpha$  is 0.521 and 0.785, for in-plane and through-plane diffusion, respectively.

The effects of saturation on the effective diffusivity of species can be generally given by power law model according to [28],

$$g(s) = (1-s)^m \tag{13}$$

As highlighted in introduction section, various researchers have used different values of exponent m.

In the present study a, relative permeability model based on percolation theory to represent the effect of saturation proposed by Dawes et al [22] has also been evaluated:

$$g(s) = \frac{\left((1-s)-0.11\right)^{0.9}}{(1-0.11)^{0.9}} \tag{14}$$

A number of auxiliary equations need to be solved to model the electrochemical reactions and determine the local current density and net water transfer coefficient. The auxiliary equations are based on the assumption of membrane Nafion 117, and are taken from the work of Springer *et al.* [4]

#### 2.4 Auxiliary Equations

The auxiliary model equations, needed to be solved to determine the net water transfer coefficient and cell voltage at average current density, are summarized below:

2.4.1 Water Transport in the membrane:

When electrochemical reaction takes place inside a fuel cell, water molecules are dragged through the membrane from the anode to the cathode with protons. This is known as electro-osmotic drag. In addition, some of the water produced at the cathode transports through the membrane from the cathode side to anode side due to concentration gradient, known as back-diffusion. Hence, the amount of water transported across the membrane is equal to the difference in water transported by the back-diffusion and that by electro-osmotic drag [17].

In order to reduce complexity in the model development, it is assumed that the water transport across the membrane is one-dimensional and can be approximated by a single step linear difference in concentrations at the cathode and anode side. The final expression for the net water transfer coefficient per proton is [9]

$$\alpha = n_d - \frac{FD_{H_2O}[C_{H_2O_c} - C_{H_2O_a}]}{It_m}$$
(15)

Where  $D_{H_2O}$  represents water diffusion coefficient, and  $C_{H_2O_a}$  and  $C_{H_2O_c}$  represent the molar concentration of water at the anode and cathode side respectively, I is the average current density and  $t_m$  is the membrane thickness and F is the Faraday's constant.

Electro-osmotic drag coefficient describes the amount of water dragged by each proton across the membrane from the anode to the cathode side and expressed as, [9]

$$n_d = 0.0049 + 2.02a_a - 4.53a_a^2 + 4.09a_a^3 ; a_a \le 1$$
  
$$n_d = 1.59 + 0.159(a_a - 1); \qquad a_a > 1$$
(16)

where, water activity is defined as, [9]

$$a_k = \frac{X_{H_{2O,K}}P}{D_{H_{2O,K}}^{sat}}$$
(17)

where P is the cell pressure and  $X_{H_2O,K}$  is the mole fraction of water on either the anode or cathode side.

Water diffusion coefficient is expressed as, [9]

$$D_{H_{20}} = 5.5e^{-11}n_d \exp[2416\left(\frac{1}{303} - \frac{1}{T}\right)]$$
(18)

Water vapour saturation pressure [9]

$$P_{H_20}^{sat} = \left[0.000644367 + 0.000213948(T - 273) + 3.4329e^{-5}(T - 273)^2 - 2.70381e^{-7}(T - 273)^3 + 8.77696e^{-9}(T - 273)^4\right] - 3.14035e^{-13}(T - 273)^5 + 3.82148e^{-14}(T - 273)^6]1.013e^{5}$$

(19)

Water concentration on the anode and cathode side, [9]

$$C_{H_2OK} = \frac{\rho_{m,dry}}{M_{m,dry}} (0.043 + 17.8a_k - 39.8a_k^2 + 36.0a_k^3; a_k \le 1$$

$$C_{H_2OK} = \frac{\rho_{m,dry}}{M_{m,dry}} (14 + 1.4(a_k - 1)); a_k > 1$$
(20)

#### 2.4.2 Polarization Characteristics

When electrical energy is drawn from the cell, the cell experiences various losses (polarization, overpotential and overvoltage losses) and a result the cell potential drops. The cell voltage can be expressed by the following equation [17]:

$$V_{\text{cell}} = E - \eta_{\text{act}} - \eta_{\text{ohm}} - \eta_{\text{conc}}$$
(21)

where E is the equilibrium thermodynamic potential which is calculated using the Nernst equation [17]:

$$E = 1.23 - 0.9 \times 10^{-3} (T - 298) + 2.3 \frac{RT}{4F} \log(p_h^2 p_0)$$
(22)

 $\eta_{act}$  is the activation overpotential,  $\eta_{ohm}$  is the ohmic overpotential and  $\eta_{conc}$  is the concentration overpotential.

Activation overpotential ( $\eta_{act}$ ):

The activation overpotential is a function of local current density, exchange current density and concentration of oxygen. The activation overpotential is expressed by Butler-Volmer equation, [17]

$$i_{a} = i_{a,ref} \left(\frac{C_{h}}{C_{h,ref}}\right)^{\frac{1}{2}} \left\{ \exp\left[\frac{\alpha_{a}n_{a}F}{RT} \eta_{act,a}\right] - \exp\left[-\frac{(1-\alpha_{a})n_{a}F}{RT} \eta_{act,a}\right] \right\}$$

$$i_{c} = i_{c,ref} \left(\frac{C_{o}}{C_{o,ref}}\right) \left\{ \exp\left[\frac{\alpha_{c}n_{c}F}{RT} \eta_{act,c}\right] - \exp\left[-\frac{(1-\alpha_{c})n_{c}F}{RT} \eta_{act,c}\right] \right\}$$
(23)

Where  $i_{a,ref}$  and  $i_{c,ref}$  are the exchange current density multiplied by specific area, n is electron number of reaction at anode or cathode and  $\alpha$  is the transfer coefficient.

Ohmic Overpotential ( $\eta_{ohm}$ ):

The ohmic overpotential occurs due to resistance to electron and ion transfer and can be expressed as [17]:

$$\eta_{\rm ohm} = \eta_{\rm ohm}^{\rm el} + \eta_{\rm ohm}^{\rm pro} = I(R^{\rm el} + R^{\rm pro})$$
(24)

Where  $R^{el}$  is the resistance to electron transfer and  $R^{pro}$  is the resistance to proton transfer. In general, the resistance to electron transfer is difficult to predict and to avoid complexity in the present model,  $R^{el} = 0.1 \Omega \text{ cm}^2$  is assumed according to the work of Min [17]. The resistance to ion transfer,  $R^{pro}$  is calculated using following expression [17]:

$$R^{pro} = \frac{t_m}{k_m} \tag{25}$$

 $t_m$  is the thickness of the membrane and  $k_m$  is the phase conductivity of the membrane. The membrane phase conductivity depends on the temperature and water concentration at anode side and expressed as

$$k_m = 100[0.00514\left(\frac{M_{m,dry}}{\rho_{m,dry}}\right)C_{H_2O_a} - 0.00326] \times \exp\left[1268(\frac{1}{303} - \frac{1}{T})\right]$$
(26)

Concentration Overpotential ( $\eta_{conc}$ ):

At high current densities, polarization losses are dominated by concentration overpotential which is caused by slow diffusion of gas phase through the porous regions. These losses can be determined by, [17]:

$$\eta_{\rm conc} = -\frac{RT}{nF} \ln \left( 1 - \frac{I}{I_{\rm L}} \right) \tag{27}$$

where  $I_{L}$  is the limiting current density [17]:

$$i_L = \frac{nFD_h C_{k,0}}{H_d} \tag{28}$$

where,  $D_h$  is the diffusion coefficient of hydrogen and  $C_{k,o}$  is the molar concentration of hydrogen before entering the gas diffusion layer,  $H_d$  is the height of the diffusion layer.

#### 2.5 Numerical procedure

The numerical methodology involves solving a set of partial differential equations of continuity, momentum, species concentrations involving oxygen, hydrogen, water vapour and liquid water. This set of equations is supplemented with auxiliary equations to take into account electro-chemical reactions. The continuity and momentum equations are solved through Ansys Fluent CFD code, while species concentration equations are solved under user defined scalar (UDS) scheme. The convection terms in the governing equations are discretised by 2<sup>nd</sup> order upwind scheme and the pressure velocity coupling is achieved by SIMPLE algorithm. The electro-chemistry has been treated explicitly with the specification of an average current density of the cell and all relevant parameters are calculated from auxiliary equations after obtaining concentration of species from their respective governing equations. The explicit electrochemistry modeling allows reducing modeling complexity of solving transport equations for voltage potential; but at the same time allows the investigation of diffusion of species through the GDL realistically, which is the main focus of this study.

#### 2.6 Computational domain and physical parameters

A representative section a three-dimensional straight channel has been considered in the present study (Figure 1). The geometry is similar to the computational work of Min [17] and Liu et al. [30]. Physical dimensions of the computational domain as well as relevant fuel cell parameters are given in Table 1. The computational domain has been meshed with quadrilateral grids of 12700 cells. A grid sensitivity test using upto 60000 cells has proved that the grid size of 12700 cells is sufficient to provide grid independency. Simulations have been carried out on a quad core Xeon workstation running on serial server. Each simulation took approximately 1000 iterations to converge in approximately 15 minutes of run time.

Dirichlet boundary conditions are applied at cathode and anode inlet. The inlet velocity is a function of stoichiometric flow ratio,  $\zeta$ , geometrical area of membrane  $A_m$  and cross-section area of gas channel,  $A_{ch}$ , reference current density,  $I_{ref}$  and concentration of reactants [30],

$$u_{c,in} = \zeta \, \frac{I_{ref}}{4 F} \frac{1}{x_{O2,in}} \frac{RT}{P} \frac{A_m}{A_{ch}} \tag{29}$$

$$u_{a,in} = \zeta \, \frac{I_{ref}}{2 F} \frac{1}{x_{H2,in}} \frac{RT}{P} \frac{A_m}{A_{ch}} \tag{30}$$

A pressure outlet condition is assumed. A symmetry boundary condition is applied on the side surfaces of the porous regions (Figure 1). No slip condition is applied to the external walls.

#### 3.0 Results and Discussions

#### 3.1 Performance Characteristics

A common practice in developing computational fuel cell model is to verify the model by comparing against V-I performance characteristics curve. Figure 2 shows the computed V-I characteristics curve for different relative diffusivity models. The figure also shows calculation from a single phase model by the authors [24] and experimental data from Ticianelli et al [31]. The V-I characteristic curves presented in Figure 2 show that the inclusion of two-phase flow modelling has significantly improved the prediction and brought the predicted cell voltage closer to the experimental data. The effect of relative diffusivity model has been predicted to be small, especially among the power law models. The model developed by Dawes et al [22] slightly overpredicts the voltage values at higher current densities. The power law model with the exponential factor of 3 is slightly closer to the experimental data; however, the results show that widely used value of exponential factor of 2 is a good approximation for relative diffusivity.

#### 3.2 Species concentrations

Further insight into the power law saturation model with exponential factor of 2 and the percolation based Dawes et al [22] model has been provided through contour plots of mass fraction of oxygen, water vapour and liquid water in the cathode side as shown in Figures 3-5. The mass fraction of oxygen drops along the flow channel as oxygen diffuses through the GDL to the catalyst layer, where oxygen is consumed. The power law and Dawes et al [22] saturation models produce very similar oxygen profile inside cathode channel, however, the Dawes et al [22] model overpredicts the

mass fraction of oxygen under the land area. Figure 4 shows the water vapour mass fraction contour; the water vapour increases downstream due to the generation of water and the depletion of oxygen. The water vapor also diffuses towards the channel through the gas diffusion layer caused by higher water vapor concentration under the land and results in curved lateral contours. Dawes et al [22] saturation model over predicts the water vapour mass fraction under the land area also. Dawes et al [22] model appears to predict higher transport of species and as a result, the Dawes et al [22] model slightly overpredicts the voltage compared to experimental data. Figure 5 shows the mass fraction contour of liquid water in the cathode side. The simulation results show that a large amount of liquid water is trapped under the land area, whereas the amount of liquid water in the channel is very small. This result highlights that the under land area is critical for water management and effective design or operating parameter should be found to remove this trapped liquid.

#### 3.3 Effects of condensation coefficient

The condensation/evaporation process of a PEM fuel cell is assumed to be governed by the partial pressure of vapor phase and temperature. However, Khan et al. [1] has questioned the validity of this assumption and highlighted that even fractional difference in temperature could have significant effects, which infers that several other unknown factors might influence the evaporation/condensation process. These unknown factors are accounted for through the condensation factor  $K_c$  in the simulation. Unfortunately, there has been no definite value for  $K_c$  and researchers have used very widely different values.

In the present study, the effects of  $K_c$  number have been investigated. Figure 6 shows the voltage current characteristics curve for  $K_c=1$  and  $K_c=10$ . The simulated cell

voltage at  $K_c = 1$  is similar to the single phase results, while the simulated results for  $K_c = 10$  agrees well with the experimental results except at very high current densities. Figures 7-9 show the contour plots of oxygen, water vapour and liquid water at the cathode side. At  $K_c = 1$ , the amount of oxygen under the land is large compared to the  $K_c = 10$  (Figures 3 and 7), whereas the amount of water vapour is almost at similar levels for the both the condensation rates (Figures 4 and 8). The contour plots of liquid water show that  $K_c = 1$  is not adequate to convert water vapour into liquid water is produced (Figure 5) compared to the Kc = 1 case. For  $K_c = 10$  case, the liquid water under the land area blocks the pores of GDL for oxygen to diffuse through and as a result the prediction with  $K_c = 10$  produces lower cell voltage and brings the prediction much closer to the experimental data.

#### 3.4 Effects of GDL permeability

The permeability of a gas diffusion layer is an important parameter that influences the performance of a fuel cell. The effects of isotropic and anisotropic permeability of a fuel cell have been investigated using the values given in Table 2. The test cases C11, C22 and C33 represent isotropic permeability. Experimental evidence suggests that in-plane permeability in gas diffusion layer is higher than through-plane permeability [19] and therefore, C11, C12, C13, C23 cases are more relevant to actual fuel cell. Simulations have been carried out for 0.5 A/cm<sup>2</sup> and 1.0 A/cm<sup>2</sup> current densities and the calculated average cell voltages have been presented in Tables 4 and 5. At 0.5 A/cm<sup>2</sup> current density, the effect of permeability is almost negligible as shown in Table 3, however, the effects of permeability becomes significant at a current density of 1.0 A/cm<sup>2</sup>. For the isotropic cases C11, C22 and C33, the decrease of permeability leads

to a large drop in fuel cell performance. For anisotropic cases C11, C12, C13, where the in-plane permeability is kept fixed at 1X10<sup>-8</sup>, and the through plane permeability has been varied 1X10<sup>-8</sup>, 1X10<sup>-10</sup> and 1X10<sup>-12</sup>, the effect is less significant with voltage dropping from 0.206 to 0.19. For the cases C11, C21, C31, where the through plane permeability is kept at a high value of 1X10<sup>-8</sup>, whereas the in-plane permeability is varied 1X10<sup>-10</sup>, 1X10<sup>-12</sup>, the effect is rather significant with the voltage decreasing from 0.206 to 0.048. The implication of this is that either the permeability of GDL should be isotropic with high enough value or anisotropic with higher in-plane permeability. The anisotropic GDL with higher through-plane permeability and lower in-plane permeability would lead to the poor performance of fuel cell. Again for C13 and C31 cases, the computed average cell voltages are 0.19 and 0.048 respectively, which clearly shows that the higher in-plane permeability compared to through plane permeability produces better cell performance.

Further insights into the effects of permeability can be gained from the profiles of mass fraction of oxygen, water vapour and liquid water at the catalyst/GDL interface at three different locations as shown in Figures 10-12. In Figure 11, the profile of mass fraction of oxygen shows two distinct groupings. For C11, C12 and C13 cases, adequate oxygen is available under the land area, but all other cases, no oxygen is available under the land area, but all other cases, no oxygen is available under the land area two possible reasons behind this: (1) less amount of oxygen is diffusing due to low permeability and (2) more importantly the presence of liquid water blocking the fresh oxygen to reach the reaction sites. Water removal rate is very good for the case of C11, C12, C13 with less amount of water (both liquid and vapour form) is present under the land area as shown in Figures 12 and 13. As a result, C11, C12 and C13 cases are the most efficient. This is also evidenced from in the cell voltage values given in Table 4.

#### 4.0 Conclusions

A two-phase computational fluid dynamics based PEM fuel cell model has been implemented to investigate the effects of diffusivity and permeability in order to determine the best way to model the transport limitation of species caused by the water flooding. A thorough comparison study of liquid water saturation model using power law with various exponential factors and a percolation based model has been reported in this study. The simulated results have been compared against the reported experimental results for characteristics curve. The present study shows that the percolation based relative permeability model overpredicts the cell voltage at higher current density, whereas the power law model with a exponential factor of 2 reproduces cell voltage much closer to the experimental data.

The effects of anisotropic and isotropic permeability have also been investigated and the simulated results show that in order to get higher fuel cell performance, the permeability of gas diffusion layer has to be high in both in-plane and through-plane directions. The higher in-plane and lower through plane permeability also produces good fuel cell performance. On the other hand, higher through-plane and lower inplane permeability produces poor fuel cell performance. As this investigation shows, this performance deterioration arises from inadequate water removal under the land area. The present study shows that by optimizing the permeability of gas diffusion higher performance of a PEM fuel cell can be obtained.

#### Nomenclature

| $a_k$ N | water activity |
|---------|----------------|
|---------|----------------|

- A specific area of the catalyst layer  $(m^{-1})$
- C molar concentration (mol m<sup>-3</sup>)
- *D* diffusion coefficient ( $m^2 s^{-1}$ )
- *E* equilibrium thermodynamic potential (V)

| F                  | Faraday constant (96485.309 C mol <sup>-1</sup> )         |
|--------------------|---|
| Н                  | Height (m)  |
| i                  | reaction rate (Am <sup>-3</sup> )                         |
| 1                  | average current density (Am <sup>-2</sup> )               |
| Κ                  | permeability (m <sup>2</sup> )                            |
| k <sub>c</sub>     | condensation rate (s <sup>-1</sup> )                      |
| k <sub>m</sub>     | phase conductivity of membrane (S m <sup>-1</sup> )       |
| L                  | length (m)  |
| Μ                  | molar mass (kg mol <sup>-1</sup> )                        |
| M <sub>m,dry</sub> | dry mass of membrane (Kg mol <sup>-1</sup> )              |
| n                  | electron number for reactions                             |
| n <sub>d</sub>     | electro-osmotic drag coefficient                          |
| Р                  | pressure (Pa)   |
| R                  | gas constant (8.314 J mol <sup>-1</sup> K <sup>-1</sup> ) |
| RH                 | relative humidity   |
| S                  | source term   |
| S                  | liquid water saturation                                   |
| t <sub>m</sub>     | membrane thickness  |
| Т                  | temperature (K)   |
| u                  | velocity vector (m s <sup>-1</sup> )                      |
| V <sub>cell</sub>  | cell voltage (V)  |
| W                  | width (m)   |
| X                  | molar fraction  |

## Greek symbols

|   | 5   |
|---|---|
| а | net water transfer coefficient                  |
| ε | porosity  |
| η | overpotential (V)                               |
| μ | viscosity (kg m <sup>-1</sup> s <sup>-1</sup> ) |
| ρ | density (kg m <sup>-3</sup> )                   |
| ω | mass fraction                                   |
| ζ | stoichiometric ratio                            |

## Subscripts and superscripts

| 0              | before diffusion layer |
|----------------|------------------------|
| а              | anode                  |
| act            | activation             |
| av             | average                |
| С              | cathode                |
| conc           | concentration          |
| d              | diffusion layer        |
| eff            | effective              |
| el             | electron               |
| H <sub>2</sub> | hydrogen               |
| k              | species                |
| L              | limiting               |
| m              | membrane               |
| O <sub>2</sub> | oxygen                 |
| ohm            | ohmic polarization     |
| pro            | proton                 |
| ref            | reference              |

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**Table 1** Physical parameters and boundary conditions used for the simulations (Min[17] and Liu [30])

| Gas channel length              | L= 100 mm                              |
|---------------------------------|--|
| Gas channel width               | W = 1 mm                               |
| Gas channel height              | $H_{ch} = 1 \text{ mm}$                |
| Diffusion layer height          | $H_{d} = 0.254 \text{ mm}$             |
| Catalyst layer height           | $H_{ct} = 0.0287 \text{ mm}$           |
| Land area width                 | $W_1 = 1 \text{ mm}$                   |
| Membrane thickness              | $t_m = 0.23 \text{ mm}$                |
| Permeability                    | $K = 1.76x \ 10^{-11} \ m^2$           |
| Faraday Constant                | $F = 96485.309 \text{ C mol}^{-1}$     |
| Operating pressure              | P = 101325 Pa                          |
| Operating temperature           | T = 323 K                              |
| GDL porosity                    | $\epsilon_{gdl} = 0.4$                 |
| CL porosity                     | $\epsilon_{cl} = 0.4$                  |
| Dry mass of membrane            | $M_{m,dry} = 1.1 \text{kgmol}^{-1}$    |
| Dry density of membrane         | $\rho_{m,dry} = 2000 \text{ kgm}^{-3}$ |
| Fuel/ air stoichiometric ratio  | $\xi_a/\xi_c = 5/5$                    |
| Electron number of anode        | $n_a = 4$                              |
| reaction                        |  |
| Electron number of cathode      | $n_c = 2$                              |
| reaction                        |  |
| Relative humidity of inlet fuel | $RH_a = 100\%$                         |
| Relative humidity of inlet air  | $RH_c = 0\%$                           |
| Oxygen mass fraction of inlet a | ir $\omega_0 = 0.232$                  |
|                                 |  |

| H <sub>2</sub> diffusion coefficient | $D_{h, ref} = 0.915 \times 10^{-4} \mathrm{m}^2 \mathrm{s}^{-1}$ |                                   |
|--------------------------------------|--|-----------------------------------|
| at reference state                   |  |                                   |
| O2 diffusion coefficient             | $D_{O, ref} = 0.22 \times 10^{-4} \mathrm{m}^2 \mathrm{s}^{-1}$  |                                   |
| at reference state                   |  |                                   |
| Water vapour diffusion coefficie     | $D_{w, ref} = 0.256 \times 10^{-5}$                              | $^{4} \text{m}^{2} \text{s}^{-1}$ |
| at reference state                   |  |                                   |
| Anode exchange current density       | $i_{a,ref} = 2.0 \times 10^8 \mathrm{Am}$                        | -3                                |
| cathode exchange current densi       | $i_{c,ref} = 160 \text{ Am}^{-3}$                                |                                   |
| Hydrogen reference concentration     | $C_{h,ref} = 56.4 \text{ mol m}^{-3}$                            | 3                                 |
| Oxygen reference concentration       | $C_{o,ref} = 3.39 \text{ mol m}^{-3}$                            | 3                                 |
| Anode transfer coefficient           | $a_{a} = 0.5$  |                                   |
| Cathode transfer coefficient         | $a_{c} = 0.5$  |                                   |
| Water vapour condensation rate       | $k_c = 10 \text{ s}^{-1}$  |                                   |

| In plane            | Through plane permeability |                     |                     |
|---------------------|----------------------------|---------------------|---------------------|
| permeability        | $1 \times 10^{-8}$         | $1 \times 10^{-10}$ | $1 \times 10^{-12}$ |
| $1 \times 10^{-8}$  | C11                        | C12                 | C13                 |
| $1 \times 10^{-10}$ | C21                        | C22                 | C23                 |
| $1 \times 10^{-12}$ | C31                        | C32                 | C33                 |

**Table 2** Combinations of permeability for the model study

**Table 3** Cell Voltage at isotropic and anisotropic conditions for average current density of 0.5 Acm-2. (porosity = 0.4)

| Case    | Cell    |
|---------|---------|
| Studies | Voltage |
| C11     | 0.583   |
| C22     | 0.571   |
| C33     | 0.568   |
| C12     | 0.584   |
| C13     | 0.577   |
| C21     | 0.571   |
| C23     | 0.571   |
| C31     | 0.568   |
| C32     | 0.568   |

**Table 4** Cell Voltage at isotropic and anisotropic conditions for average current density of 1  $Acm^{-2}$ .(GDL and CL porosity = 0.4)

| Case    | Cell    |
|---------|---------|
| Studies | Voltage |
| C11     | 0.206   |
| C22     | 0.156   |
| C33     | 0.026   |
| C12     | 0.207   |
| C13     | 0.190   |
| C21     | 0.159   |
| C23     | 0.148   |
| C31     | 0.048   |
| C32     | 0.052   |

Figure 1: Schematic diagram of the three-dimensional PEM fuel cell model







**Figure 3:** Contour plot of mass fraction of oxygen at the cathode side. (a) Power Law model, (1-S)<sup>2</sup> and (b) Percolation model, Dawes et al [12]



(b)





(a)



(b)

**Figure 5:** Contour plot of mass fraction of liquid water at the cathode side. (a) Power Law model, (1-S)<sup>2</sup> and (b) Percolation model, Dawes et al [12].





(b)

Figure 6: Comparison of the coefficient of condensation for predicting V-I Characteristics curve.



Figure 7: Contour plot of mass fraction of oxygen at the cathode side. (a) Power Law model,  $(1-S)^2$  with  $K_c=1$ .



Figure 8: Contour plot of mass fraction of water vapour at the cathode side. (a) Power

Law model,  $(1-S)^2$  with  $K_c=1$ .



Figure 9: Contour plot of mass fraction of liquid water at the cathode side. (a) Power Law model,  $(1-S)^2$  with  $K_c=1$ .









0.04



Figure 11: Mass fraction of water vapour for different permeability cases at the GDL-Catalyst interface: (a) inlet, (b) middle (c) outlet sections.

(c) outlet



Figure 12: Mass fraction of water vapour for different permeability cases at the GDL-Catalyst interface: (a) inlet, (b) middle (c) outlet sections.

(c) outlet