Modeling optimizes PEM fuel cell performance using three-dimensional multi-phase computational fluid dynamics model

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Abstract

This paper presents the results of an optimization study using a comprehensive three-dimensional, multi-phase, non-isothermal model of a PEM fuel cell that incorporates the significant physical processes and the key parameters affecting fuel cell performance. The model accounts for both the gas and liquid phase in the same computational domain and, thus, allows for the implementation of phase change inside the gas diffusion layers. The model includes the transport of gaseous species, liquid water, protons, energy and water dissolved in the ion conducting polymer. Water is assumed to be exchanged among three phases; liquid, vapor and dissolved, and equilibrium among these phases is assumed. The model features an algorithm that allows a more realistic representation of the local activation overpotentials, which leads to improved prediction of the local current density distribution. This model also takes 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 results showed that the present multi-phase model is capable of identifying important parameters for the wetting behavior of the gas diffusion layers and can be used to identify conditions that might lead to the onset of pore plugging, which has a detrimental effect on the fuel cell performance. This model is used to study the effects of several operating, design and material parameters on fuel cell performance. Detailed analyses of the fuel cell performance under various operating conditions have been conducted and examined.

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1. Introduction

Water management is one of the critical operation issues in proton exchange membrane (PEM) fuel cells. Spatially varying concentrations of water in both vapor and liquid form are expected throughout the cell because of varying rates of production and transport [1]. Devising better water management is, therefore, a key issue in PEMFC design, and this requires improved understanding of the parameters affecting water transport in the membrane. Thermal management is also required to remove the heat produced by the electrochemical reaction in order to prevent drying out the membrane, which, in turn, can result not only in reduced performance but also in eventual rupture of the membrane [2]. The small temperature differentials between the fuel cell stack and the operating environment make thermal management a challenging problem in PEM fuel cells. Thermal management is also essential for control of the water evaporation or condensation rates.

The difficult experimental environment of fuel cell systems has stimulated efforts to develop models that could simulate and predict multi-dimensional coupled transport of reactants, heat and charged species using computational fluid dynamic (CFD) methods. A comprehensive computational model should include the equations and other numerical relations needed to define fully the fuel cell behavior over the range of interest. Early multi-dimensional models described gas transport in the flow channels,
gas diffusion layers and the membrane [3–5]. Recently, there has been an interest in describing operating regimes that are dominated by mass transport limitations, resulting, in part, from the formation and transport of liquid water within the fuel cell. To model fuel cell performance in these regimes, it is necessary to include equations that describe not only the motion of water within the liquid within the fuel cell. To model fuel cell performance in these regimes, it is necessary to include equations that describe not only the motion of water within the liquid water concentration. This model provides information on liquid water saturation and flooding under various conditions, but it does not account for water dissolved in the ion conducting polymer to calculate water content through the membrane.

Hu et al. [8,9] developed an isothermal, three-dimensional, two phase model for a PEM fuel cell. Their model accounts for the transport of liquid water within the porous electrodes and water dissolved in the ion conducting polymer. In addition to being restricted to constant cell temperature, the simulations presented so far using the mixture model are restricted to low humidity inlet feed streams.

Simulations of 3D-PEM fuel cell geometries have required some simplifications in order to reduce computational requirements. In particular, it is only very recently that those CFD based models have started to account for non-uniform distributions of overpotentials at the electrodes [10,11]. These models provide comprehensive details on the distribution of reactants and allow sensitivity analysis and prediction of performance under various conditions, but they do not account for phase change, liquid water concentration in the electrodes and water dissolved

Siegel et al. [6] developed a multi-phase, two-dimensional model to take into account the liquid water saturation and flooding effect, and they have studied transport limitations due to water build up in the cathode catalyst region. In a real PEM fuel cell geometry, the gas diffusion layers are used to enhance the reaction area accessible by the reactants. The effect of using these diffusion layers is to allow a spatial distribution in the current density on the membrane in both the direction of bulk flow and the direction orthogonal to the flow but parallel to the membrane. This two-dimensional distribution cannot be modeled with the well used two-dimensional models where the mass transport limitation is absent in the third direction.

Berning and Djilali [7] developed a computational fluid dynamics multi-phase model of a PEM fuel cell. Their model accounts for three-dimensional transport processes including phase change and heat transfer and including
in the ion conducting polymer. A common issue in many computational models is the uncertainties associated with the specification of various parameters that impact the transport processes. Assessing the sensitivity of the flow and the thermal or electrochemical response to these uncertainties is essential.

The development of physically representative models that allow reliable simulation of the processes under realistic conditions is essential to the development and optimization of fuel cells, the introduction of cheaper materials and fabrication techniques and the design and development of novel architectures. Recently, Wang et al. [12] and Berning and Djilali [4] performed a parametric study using a single-phase three-dimensional model. The impact of liquid water on transport in the gas diffusion electrode was, however, not taken into account. Liquid water impacts are inherently parametric, but no studies have yet been published to address this.

2. Model description

The present work presents a comprehensive three-dimensional, multi-phase, non-isothermal model of a PEM fuel cell that incorporates the significant physical processes and the key parameters affecting fuel cell performance. The model accounts for both gas and liquid phases in the same computational domain and, thus, allows implementation of phase change inside the gas diffusion layers. The model includes the transport of gaseous species, liquid water, protons, energy and water dissolved in the ion conducting polymer. Water transport inside the porous gas diffusion layer and catalyst layer is described by two physical mechanisms: viscous drag and capillary pressure forces, and is described by advection within the gas channels. Water transport across the membrane is also described by two physical mechanisms: electro-osmotic drag and diffusion. Water is assumed to be exchanged among three phases; liquid, vapor and dissolved, and equilibrium among these phases is assumed. The model features an algorithm that allows for a more realistic representation of the local activation overpotentials, which leads to improved prediction of the local current density distribution. This model also takes 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 model reflects the influence of numerous parameters on fuel cell performance including geometry, materials, operating and others. The present multi-phase model is capable of identifying important parameters for the wetting behavior of the gas diffusion layers and can be used to identify conditions that might lead to the onset of pore plugging, which has a detrimental effect on the fuel cell performance. This paper describes 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. An optimization study using this model has been performed. The study quantifies the impact of operating, design and material parameters on fuel cell performance.

2.1. Computational domain

A computational model of an entire cell would require very large computing resources and excessively long simulation times. The computational domain in this study is, therefore, limited to one straight flow channel with the land areas. The full computational domain consists of cathode and anode gas flow channels and the membrane electrode assembly as shown in Fig. 1.

2.2. Model equations

2.2.1. Gas flow channels

In the fuel cell channels, the gas flow field is obtained by solving the steady state Navier–Stokes equations, i.e. the continuity equation, along with the momentum equations and the mass conservation equation for each phase yields the volume fraction \( r \) and the pressure distribution inside the channels. The continuity equation for the gas phase inside the channel is given by

\[
\nabla \cdot (r \rho_g u_g) = 0
\]

and for the liquid phase inside the channel

\[
\nabla \cdot (r \rho_l u_l) = 0
\]

Two sets of momentum equations are solved in the channels, and they share the same pressure field. Under these conditions, it can be shown that the momentum equations become

![Fig. 1. Three-dimensional computational domain.](image-url)
\[ \nabla \cdot (\rho_g \mathbf{u}_g \otimes \mathbf{u}_g - \mu_g \nabla \mathbf{u}_g) = -\nabla r_g \left( P + \frac{2}{3} \mu_g \frac{\nabla \cdot \mathbf{u}_g}{\mu_g} \right) + \nabla \cdot \left[ \mu_g (\nabla \mathbf{u}_g)^T \right] \] (3)

and

\[ \nabla \cdot (\rho_i \mathbf{u}_i \otimes \mathbf{u}_i - \mu_i \nabla \mathbf{u}_i) = -\nabla r_i \left( P + \frac{2}{3} \mu_i \frac{\nabla \cdot \mathbf{u}_i}{\mu_i} \right) + \nabla \cdot \left[ \mu_i (\nabla \mathbf{u}_i)^T \right] \] (4)

The mass balance is described by the divergence of the mass flux through diffusion and convection. Multiple species are considered in the gas phase only, and the species conservation equation in multi-component, multi-phase flow can be written in the following expression for species \( i \):

\[
\nabla \cdot \left[ -r_g \rho_g y_j \sum_{j=1}^N D_{ij} \frac{M_j}{M} \left( \nabla y_j + y_j \frac{\nabla M_j}{M} \right) \right] + (x_j - y_j) \frac{\nabla P}{P} + r_g \rho_g y_i \mathbf{u}_g + D_{ij} \frac{\nabla T}{T} = 0
\] (5)

where the subscript \( i \) denotes oxygen at the cathode side and hydrogen at the anode side and \( j \) is water vapor in both cases. Nitrogen is the third species at the cathode side.

The Maxwell–Stefan diffusion coefficients of any two species are dependent on temperature and pressure. They can be calculated according to the following empirical relation based on kinetic gas theory [13]:

\[
D_{ij} = \frac{T^{1.75} \times 10^{-3}}{P \left[ \left( \sum k V_{ki} \right)^{1/3} + \left( \sum k V_{kj} \right)^{1/3} \right]^2} \left[ \frac{1}{M_i} + \frac{1}{M_j} \right]^{1/2}
\] (6)

In this equation, pressure is in [atm], and the binary diffusion coefficient is in [cm²/s]. The values for \( \left( \sum k V_{ki} \right) \) are given by Fuller et al. [13].

The temperature field is obtained by solving the convective energy equation:

\[
\nabla \cdot \left( r_g \rho_g C_p \mathbf{u}_g (T - k_g \nabla T) \right) = 0
\] (7)

The gas phase and the liquid phase are assumed to be in thermodynamic equilibrium; hence the temperature of the liquid water is the same as the gas phase temperature.

### 2.2.2. Gas diffusion layers (GDL)

The physics of multiple phases through a porous medium is further complicated here by phase change and the sources and sinks associated with the electrochemical reaction. The equations used to describe transport in the gas diffusion layers are given below.

Mass transfer in the form of evaporation \( (\dot{m}_{\text{phase}} > 0) \) and condensation \( (\dot{m}_{\text{phase}} < 0) \) is assumed, so the mass balance equations for both phases are

\[
\nabla \cdot (\left( 1 - \text{sat} \right) \rho_g C_p \mathbf{u}_g) = \dot{m}_{\text{phase}}
\] (8)

and

\[
\nabla \cdot \left( \text{sat} \cdot \rho_l \mathbf{u}_k \right) = \dot{m}_{\text{phase}}
\] (9)

The momentum equation for the gas phase reduces to Darcy’s law, which is, however, based on the relative permeability for the gas phase \( (KP) \). The relative permeability accounts for the reduction in pore space available for one phase due to the existence of the second phase [7].

The momentum equation for the gas phase inside the gas diffusion layer becomes

\[
\mathbf{u}_g = -(1 - \text{sat}) \frac{KP}{\mu_g} \nabla P
\] (10)

Two liquid water transport mechanisms are considered; shear, which drags the liquid phase along with the gas phase in the direction of the pressure gradient, and capillary forces, which drive liquid water from high to low saturation regions [7]. Therefore, the momentum equation for the liquid phase inside the gas diffusion layer becomes

\[
\mathbf{u}_l = -\frac{KP_l}{\mu_l} \nabla P + \frac{K P_\text{sat}}{\mu_\text{sat}} \frac{\partial P_\text{sat}}{\partial \text{sat}} \nabla \text{sat}
\]

The functional variation of capillary pressure with saturation is prescribed following Leverett [7] who has shown that

\[
P_\text{sat} = \sigma \left( \frac{\kappa}{KP} \right)^{1/2} (1.417(1 - \text{sat}) - 2.12(1 - \text{sat})^3 + 1.263(1 - \text{sat})^3)
\] (11)

The liquid phase consists of pure water, while the gas phase has multi-components. The transport of each species in the gas phase is governed by a general convection–diffusion equation in conjunction with the Stefan–Maxwell equations to take into account multi-species diffusion:

\[
\nabla \cdot \left[ -(1 - \text{sat}) \rho_g y_j \sum_{j=1}^N D_{ij} \frac{M_j}{M} \left( \nabla y_j + y_j \frac{\nabla M_j}{M} \right) \right] + (x_j - y_j) \frac{\nabla P}{P} + (\dot{m}_{\text{phase}}/T) = 0
\] (13)

In order to account for geometric constraints of the porous media, the diffusivities are corrected using the Bruggemann correction formula [10]:

\[
D_{ij}^{\text{eff}} = D_{ij} \times \left( \frac{\varepsilon}{\varepsilon + 1} \right)^{1.5}
\] (14)

The heat transfer in the gas diffusion layers is governed by the energy equation as follows:

\[
\nabla \cdot ((1 - \text{sat}) \rho_g C_p \mathbf{u}_g T - k_g \mathbf{u}_g \nabla T) = \varepsilon \beta (T_{\text{solid}} - T) - \dot{m}_{\text{phase}} \Delta H_{\text{evap}}
\] (15)

where the term \( \varepsilon \beta (T_{\text{solid}} - T) \), on the right hand side, accounts for the heat exchange to and from the solid matrix of the GDL. \( \beta \) is a modified heat transfer coefficient that accounts for the convective heat transfer in [W/m²] and the specific surface area [m²/m³] of the porous medium [3]. Hence, the unit of \( \beta \) is [W/m³].
The gas phase and the liquid phase are assumed to be in thermodynamic equilibrium, i.e. the liquid water and the gas phase are at the same temperature.

The potential distribution in the gas diffusion layers is governed by
\[ \nabla \cdot (\lambda_s \nabla \phi) = 0 \]  
In order to account for the magnitude of phase change inside the GDL, expressions are required to relate the levels of over and under saturation as well as the amount of liquid water present to the amount of water undergoing phase change. In the present work, the procedure of Berning and Djilali [7] was used to account for the magnitude of phase change inside the GDL as follows.

In the case of evaporation, such relations must be dependent on (i) the level of under saturation of the gas phase in each control volume and on (ii) the surface area of the liquid water in the control volume. The surface area can be assumed proportional to the volume fraction of the liquid water in each cell. A plausible choice for the shape of the liquid water is droplets, especially since the catalyst area is teflonated [7].

The evaporation rate of a droplet in a convective stream depends on the rate of under saturation, the surface area of the liquid droplet and a (diffusivity dependent) mass transfer coefficient. The mass flux of water undergoing evaporation in each control volume can be represented by [7]
\[ \dot{m}_{\text{evap}} = M_{H_2O} \sigma N_p k_{\text{sm}} \pi D_{\text{drop}} \frac{x_{w0} - x_{w_{\text{wc}}}}{1 - x_{w0}} \]  

The bulk concentration \( x_{w_{\text{wc}}} \) is known by solving the continuity equation of the water vapor. To obtain the concentration at the surface \( x_{w0} \), it is reasonable to assume thermodynamic equilibrium between the liquid phase and the gas phase at the interface, i.e. the relative humidity of the gas in the immediate vicinity of the liquid is 100%. Under that condition, the surface concentration can be calculated based on the saturation pressure and is only a function of temperature.

The heat transfer coefficient for convection around a sphere is well established, and by invoking the analogy between convective heat and mass transfer, the following mass transfer coefficient was obtained by Berning and Djilali [7]:
\[ k_{\text{sm}} = \frac{c_{\text{air}} D_{H_2O}}{D_{\text{drop}}} \left[ 2 + 0.6 \left( \frac{D_{\text{drop}} v_{\infty} \rho_g}{\mu_g} \right)^{1/2} \left( \frac{\mu_g}{\rho_g D_{H_2O}} \right)^{1/3} \right] \]

It is further assumed that all droplets have a specified diameter \( D_{\text{drop}} \), and the number of droplets in each control volume is found by dividing the total volume of the liquid phase in each control volume by the volume of one droplet:
\[ N_D = \frac{\text{sat} \cdot V_{cy}}{\frac{1}{6} \pi D_{\text{drop}}} \]  

In the case when the calculated relative humidity in a control volume exceeds 100%, condensation occurs, and the evaporation term is switched off.

The case of condensation is more complex, because it can occur on every solid surface area, but the rate of condensation can be different when it takes place on a wetted surface. In addition, the overall surface area in each control volume available for condensation shrinks with an increasing amount of liquid water present. Berning and Djilali [7] assumed that the rate of condensation depends only on the level of over saturation of the gas phase multiplied by a condensation constant. Thus, the mass flux of water undergoing condensation in each control volume can be represented by
\[ \dot{m}_{\text{cond}} = \frac{\sigma C x_{w0} - x_{w_{\text{wc}}}}{1 - x_{w0}} \]  

2.2.3. Catalyst layers

The catalyst layer is treated as a thin interface, where sink and source terms for the reactants are implemented. Because of the infinitesimal thickness, the source terms are actually implemented in the last grid cell of the porous medium. At the cathode side, the sink term for oxygen is given by
\[ S_{O_2} = -\frac{M_{O_2}}{4F} i_c \]  
whereas the sink term for hydrogen is specified as
\[ S_{H_2} = -\frac{M_{H_2}}{2F} i_c \]  

The production of water is modeled as a source term and, hence, can be written as
\[ S_{H_2O} = \frac{M_{H_2O}}{2F} i_c \]  

The generation of heat in the cell is due to entropy changes as well as irreversibilities due to the activation overpotential [14]:
\[ \dot{q} = \frac{\left[ T(-\Delta s) \right]}{n_c F} + \eta_{\text{act,}c} i_c \]

The local current density distribution in the catalyst layers is modeled by the Butler–Volmer equation [12,15]:
\[ i_c = \frac{\eta_{\text{ref}}}{\eta_{\text{act},c}} \left( \frac{C_{O_2}}{\text{ref}} \right) \exp \left( \frac{\alpha_F F}{RT} \eta_{\text{act},c} \right) + \exp \left( -\frac{\alpha_F F}{RT} \eta_{\text{act},c} \right) \]
\[ i_a = \frac{\eta_{\text{ref}}}{\eta_{\text{act},a}} \left( \frac{C_{H_2}}{\text{ref}} \right)^{1/2} \left[ \exp \left( \frac{\alpha_F F}{RT} \eta_{\text{act},a} \right) + \exp \left( -\frac{\alpha_F F}{RT} \eta_{\text{act},a} \right) \right] \]

2.2.4. Membrane

The balance between the electro-osmotic drag of water from the anode to cathode and back diffusion from the cathode to anode yields the net water flux through the membrane:
The water diffusivity in the polymer can be calculated as follows [6]:

\[
D_W = 1.3 \times 10^{-10} \exp \left( \frac{2416}{303} - \frac{1}{7} \right)
\]  

(28)

The variable \( c_w \) represents the number of water molecules per sulfonic acid group (i.e. \( \text{mol H}_2\text{O/equivalent SO}_3^- \)). The water content in the electrolyte phase is related to the water vapor activity via [8,9]:

\[
c_w = 0.043 + 17.81a - 39.85a^2 + 36.0a^3 \quad (0 < a < 1)
\]

(29)

\[
c_w = 14.0 + 1.4(a - 1) \quad (1 < a < 3)
\]

(30)

\[c_w = 16.8 \quad (a > 3)\]

The water vapor activity is given by

\[
W = \frac{x_w \rho}{x_w \rho_{\text{sat}}}
\]

(31)

Heat transfer in the membrane is governed by

\[
\nabla \cdot (k_{\text{mem}} \nabla T) = 0
\]

(32)

2.3. Boundary conditions

Boundary conditions have to be applied for all variables of interest in the computational domain. In order to reduce computational cost, advantage is taken of the geometric periodicity of the cell. Hence, symmetry is assumed in the y-direction, i.e. all gradients in the y-direction are set to zero at the x-z plane boundaries of the domain. With the exception of the channel inlets and outlets, a zero flux condition is applied at all x-boundaries (x-planes).

The inlet values at the anode and cathode are prescribed for the velocity, temperature and species concentrations (Dirichlet boundary conditions). The gas streams entering the cell are fully humidified, but no liquid water is contained in the gas stream. The inlet velocities of air and fuel are calculated based on the desired current density according to

\[
u_{\text{in,c}} = \frac{L_{\text{MEA}}}{A_{\text{ch}}} \frac{1}{\rho_{\text{c,in}}} \frac{RT_{\text{in,c}}}{P_{\text{c,in}}} \frac{1}{A_{\text{ch}}}
\]

(33)

\[
u_{\text{in,a}} = \frac{L_{\text{MEA}}}{A_{\text{ch}}} \frac{1}{\rho_{\text{a,in}}} \frac{RT_{\text{in,a}}}{P_{\text{a,in}}} \frac{1}{A_{\text{ch}}}
\]

(34)

At the outlets of the gas flow channels, only the pressure is being prescribed as the desired electrode pressure; for all other variables, the gradient in the flow direction (x) is assumed to be zero (Neumann boundary conditions).

At the external surfaces in the z-direction, (top and bottom surfaces of the cell), temperature is specified and zero heat flux is applied at the x-y plane of the conducting boundary surfaces. Combinations of Dirichlet and Neumann boundary conditions are used to solve the electronic and protonic potential equations. Dirichlet boundary conditions are applied at the anode and cathode sides. Neumann boundary conditions are applied at the land area. Neumann boundary conditions are applied at the interface between the gas channels and the gas diffusion layers to give zero potential flux into the gas channels. Similarly, the protonic potential field requires a set of potential boundary conditions and zero flux boundary condition at the anode catalyst layer interface and cathode catalyst layer interface, respectively.

3. Solution algorithm

The governing equations were discretized 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 solution begins by specifying a desired current density of the cell to use for calculating the inlet flow rates at the anode and cathode sides. An initial guess of the activation overpotential is obtained from the desired current density using the
Butler–Volmer equation. Then computing the flow fields for each phase for velocities \( u, v, w \) and pressure \( P \) follows. Once the flow field is obtained, the mass fraction equations are solved for the mass fractions of oxygen, hydrogen, nitrogen and water. Scalar equations are solved last in the sequence of the transport equations for the temperature field in the cell and potential fields in the gas diffusion layers and the membrane. The local current densities are solved based on the Butler–Volmer equation. After the local current densities are obtained, the local activation overpotentials can be readily calculated from the Butler–Volmer equation. The local activation overpotentials are updated after each global iterative loop. Convergence criteria are then applied on each variable, and the procedure is repeated until convergence. The properties are updated after each global iterative loop based on the new local gas composition and temperature. Source terms reflect changes in the overall gas phase mass flow due to consumption or production of gas species via reaction and due to mass transfer between water in the vapor phase and water that is in the liquid phase or dissolved in the polymer (phase change).

The strength of the current model is clearly to perform parametric studies and explore the impact of various parameters on the transport mechanisms and on fuel cell performance. The feature of the algorithm developed in this work is its capability for accurate calculation of the local activation overpotentials, which, in turn, results in improved prediction of the local current density distribution. The flow diagram of the algorithm is shown in Fig. 2.

4. Results and discussion

The multi-phase model is validated by comparing the model results to experimental data provided by Wang et al. [12]. The values of the electrochemical transport parameters are tabulated in Table 1.

### Table 1: Electrode and membrane parameters for base case operating conditions

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrode porosity</td>
<td>( \varepsilon )</td>
<td>0.4</td>
<td>–</td>
<td>[16]</td>
</tr>
<tr>
<td>Electrode electronic conductivity</td>
<td>( \lambda_e )</td>
<td>100</td>
<td>S/m</td>
<td>[11]</td>
</tr>
<tr>
<td>Membrane ionic conductivity</td>
<td>( \lambda_m )</td>
<td>17.1223</td>
<td>S/m</td>
<td>[16]</td>
</tr>
<tr>
<td>Transfer coefficient, anode side</td>
<td>( \alpha_a )</td>
<td>0.5</td>
<td>–</td>
<td>[17]</td>
</tr>
<tr>
<td>Transfer coefficient, cathode side</td>
<td>( \alpha_c )</td>
<td>1</td>
<td>–</td>
<td>[15]</td>
</tr>
<tr>
<td>Cathode reference exchange current density</td>
<td>( \iota_{ref} )</td>
<td>1.8081e–3</td>
<td>A/m²</td>
<td>[4]</td>
</tr>
<tr>
<td>Anode reference exchange current density</td>
<td>( \iota_{ref} _a )</td>
<td>2465.598</td>
<td>A/m²</td>
<td>[4]</td>
</tr>
<tr>
<td>Electrode thermal conductivity</td>
<td>( k_{eff} )</td>
<td>1.3</td>
<td>W/m²K</td>
<td>[10]</td>
</tr>
<tr>
<td>Membrane thermal conductivity</td>
<td>( k_{mem} )</td>
<td>0.455</td>
<td>W/m²K</td>
<td>[10]</td>
</tr>
<tr>
<td>Electrode hydraulic permeability</td>
<td>( k_p )</td>
<td>1.76e–11</td>
<td>m²/s</td>
<td>[12]</td>
</tr>
<tr>
<td>Entropy change of cathode side reaction</td>
<td>( \Delta S )</td>
<td>–326.36</td>
<td>J/mole K</td>
<td>[14]</td>
</tr>
<tr>
<td>Heat transfer coefficient between solid and gas phase</td>
<td>( \beta )</td>
<td>4e6</td>
<td>W/m³</td>
<td>[3]</td>
</tr>
<tr>
<td>Protonic diffusion coefficient</td>
<td>( D_H^+ )</td>
<td>4.5e–9</td>
<td>m²/s</td>
<td>[16]</td>
</tr>
<tr>
<td>Fixed-charge concentration</td>
<td>( \epsilon_f )</td>
<td>2465.598</td>
<td>mole/m³</td>
<td>[16]</td>
</tr>
<tr>
<td>Fixed-site charge</td>
<td>( z_f )</td>
<td>1</td>
<td>–</td>
<td>[16]</td>
</tr>
<tr>
<td>Electro-osmotic drag coefficient</td>
<td>( n_d )</td>
<td>2.5</td>
<td>–</td>
<td>[18]</td>
</tr>
<tr>
<td>Droplet diameter</td>
<td>( D_{drop} )</td>
<td>1.0 × 10⁻⁸</td>
<td>m</td>
<td>[7]</td>
</tr>
<tr>
<td>Condensation constant</td>
<td>( C )</td>
<td>1.0 × 10⁻⁵</td>
<td>–</td>
<td>[7]</td>
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<tr>
<td>Scaling parameter for evaporation</td>
<td>( m )</td>
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<td>[7]</td>
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### Table 2: Geometrical and operational parameters for base case conditions

<table>
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<th>Parameter</th>
<th>Symbol</th>
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<th>Unit</th>
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<td>Channel length</td>
<td>( L )</td>
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<td>m</td>
</tr>
<tr>
<td>Channel width</td>
<td>( W )</td>
<td>1e–3</td>
<td>m</td>
</tr>
<tr>
<td>Channel height</td>
<td>( H )</td>
<td>1e–3</td>
<td>m</td>
</tr>
<tr>
<td>Land area width</td>
<td>( W_{land} )</td>
<td>1e–3</td>
<td>m</td>
</tr>
<tr>
<td>Gas diffusion layer thickness</td>
<td>( \delta_{GDL} )</td>
<td>0.26e–3</td>
<td>m</td>
</tr>
<tr>
<td>Wet membrane thickness (Nafion® 117)</td>
<td>( \delta_{mem} )</td>
<td>0.23e–3</td>
<td>m</td>
</tr>
<tr>
<td>Catalyst layer thickness</td>
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<td>Hydrogen reference mole fraction</td>
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<td>–</td>
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<tr>
<td>Oxygen reference mole fraction</td>
<td>( x_{O_2}^{ref} )</td>
<td>0.17774</td>
<td>–</td>
</tr>
<tr>
<td>Anode pressure</td>
<td>( P_a )</td>
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<td>atm</td>
</tr>
<tr>
<td>Cathode pressure</td>
<td>( P_c )</td>
<td>3</td>
<td>atm</td>
</tr>
<tr>
<td>Inlet fuel and air temperature</td>
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<td>K</td>
</tr>
<tr>
<td>Relative humidity of inlet fuel and air (fully humidified conditions)</td>
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<td>100</td>
<td>%</td>
</tr>
<tr>
<td>Air stoichiometric ratio</td>
<td>( \xi_e )</td>
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<tr>
<td>Fuel stoichiometric ratio</td>
<td>( \xi_a )</td>
<td>2</td>
<td>–</td>
</tr>
</tbody>
</table>

Fig. 2. Flow diagram of the solution procedure used.
parameters for the base case operating conditions are listed in Table 1. The geometric and the base case operating conditions are listed in Table 2. It is important to note that because this model accounts for all major transport processes and the modeling domain comprises all the elements of a complete cell, no parameters needed to be adjusted in order to obtain physical results.

The importance of phase change to accurate modeling of fuel cell performance is illustrated. Performance curves with and without phase change as well as experimental data are shown in Fig. 3 for the base case conditions. Comparison of the two curves demonstrates that the effects of liquid water accumulation become apparent even at relatively low values of current density. Furthermore, when liquid water effects are not included in the model, the cell voltage does not exhibit an increasingly steep drop as the cell approaches its limiting current density. This drop off in performance is clearly demonstrated by experimental data but cannot be accurately modeled without incorporation of phase change. By including the effects of phase change, the current model is able to simulate more closely the performance, especially in the region where mass transport effects begin to dominate.

The performance characteristics of the fuel cell based on a certain parameter can be obtained by varying that parameter while keeping all other parameters constant. Results obtained from these parametric studies will allow us to identify the critical parameters for fuel cell performance. Results with different operating conditions for the cell operating at nominal current density of 1.2 A/cm² are discussed in the following sections.

4.1. Effect of temperature

The temperature basically affects all the different transport phenomena inside the fuel cell. The composition of the incoming gas streams depends strongly on the temp-

![Fig. 3. Comparison of three-dimensional simulation with an experimental polarization curve.](image)

![Fig. 4. Temperature profiles along through plane direction for three different cell operating temperatures of 60 °C, 80 °C and 90 °C.](image)
perature. Assuming the inlet gases are fully humidified, the partial pressure of water vapor entering the cell depends on the temperature only. Thus, the molar fraction of water vapor is a function of the total inlet pressure and temperature, and so, the molar fraction of incoming hydrogen and oxygen depend on the temperature and pressure as well. At an operating pressure of 1 atm, the effect of the temperature on the inlet composition is much stronger than at elevated pressures. At 90 °C, for atmospheric pressure, almost 68% (molar) of the incoming cathode side gas stream consists of water vapor, and only around 6.6% is oxygen. The result is a decrease in the molar oxygen fraction at the catalyst layer. It is expected that this will affect mainly the limiting current density. The molar oxygen fraction at the catalyst layer decreases with increasing cell operating temperature due to the reduction of the molar oxygen fraction in the incoming gas streams. The result is a much higher fraction of the local current density, and it is generated under the channel area. Fig. 4 shows the temperature profiles in the through plane direction where data is taken at $x = 10$ mm length of the cell cutting across the middle of the flow channel. The results show that the temperature distribution inside the cell shows that the maximum temperature occurs at the higher cell operating temperature case (90 °C), but the maximum temperature gradient ($\Delta T$) appears in the lower cell operating temperature case (60 °C) due to the high activation overpotential, which leads to more heat generation with the drop of cell voltage. The activation overpotential
decreases with increasing cell operating temperature. This is because the exchange current density of the oxygen reduction reaction increases rapidly with temperature due to the enhanced reaction kinetics, which reduces the activation losses. A higher temperature leads also to a higher diffusivity of the hydrogen protons in the electrolyte membrane, thereby reducing the membrane resistance, and this leads to reducing the potential loss in the membrane. The mass transport loss increases as the cell operating temperature increases due to the reduction of the molar oxygen fraction in the incoming gas streams and, hence, a reduction in the molar oxygen fraction at the catalyst layer.

4.2. Effect of pressure

Similar to the temperature, the operating pressure affects numerous transport parameters that are important for the fuel cell operation. The saturation pressure of water vapor depends only on the temperature, and it remains constant for a variation of the inlet pressure. A change in the operating pressure leads to a change in the inlet gas compositions, assuming the inlet gases are fully humidified. The increase in the molar oxygen fraction in the incoming gas streams is significant when the pressure is increased from atmospheric pressure up to 3 atm. A further increase in the pressure from 3 atm to 5 atm does not lead to a signif-

Fig. 6. Temperature distribution at the cathode side catalyst layer for two different values of the stoichiometric flow ratio: (a) 1.5 and (b) 3.
significant improvement in terms of the molar oxygen fraction. The effect of the cell operating pressure on the local current distribution shows that the higher cell operating pressure results in more even distribution of the local current density due to the high oxygen concentration at the catalyst layer. This leads to the fact that for a lower cell operating pressure at a constant nominal current density, there is a much stronger distribution of current inside the cell, the maximum local current density being at the inlet under the channel area. Therefore, the maximum temperature gradient appears in the cathode side catalyst layer of the lower cell operating pressure as can be seen in Fig. 5. The activation overpotential decreases with increasing cell operating pressure. This is because the exchange current density of the oxygen reduction reaction increases with increasing cell operating pressure due to the enhanced reaction kinetics. To reduce mass transport loss, the cathode is usually run at high pressure. In essence, higher pressures help to force the oxygen and hydrogen into contact with the electrolyte, and this leads to reducing the mass transport loss.

4.3. Effect of stoichiometric flow ratio

The stoichiometric flow ratio has an important impact on the water management in the cell. The amount of

Fig. 7. Temperature distribution at the cathode side catalyst layer for two different values of the channel width: (a) 0.8 mm and (b) 1.2 mm.
incoming air determines how much water vapor can be carried out of the cell. The effect of the stoichiometric flow ratio on the local current distribution shows that the higher stoichiometric flow ratio results in more even distribution of the local current density due to the high oxygen concentration at the catalyst layer. Therefore, the maximum temperature gradient appears in the cathode side catalyst layer of the lower stoichiometric flow ratio case as can be seen in Fig. 6. For an optimum fuel cell performance, and in order to avoid large temperature gradients inside the fuel cell, it is desirable to achieve a uniform current density distribution inside the cell.

### 4.4. Effect of gas channels width

A reduction in the land area width by increasing the width of the gas flow channel enhances the mass transport of the reactions to the catalyst layer that lies under the land area. The result is an increase in the molar oxygen fraction at the catalyst layer with more even distribution. It is expected that this will affect mainly the limiting current density and, to a lesser degree, the voltage drop due to mass transport limitations. The channel width has a large impact on the local current density distribution. For the narrow channel, the local current density can exceed more than 40% of the nominal current density with a sharp drop.

![Temperature distribution at the cathode side catalyst layer for two different GDL thicknesses: (a) 0.2 mm and (b) 0.3 mm.](image-url)
off under the land area, where the local current density is about 40% lower than the nominal current density. The wider channel makes a much more evenly distributed current throughout the cell. However, the temperature peak appears in the cathode side catalyst layer of the wider channel case as can be seen in Fig. 7, implying that the major heat generation takes place where the electrochemical activity is highest. This is because the increase in the width of the gas flow channel means that the velocity of the incoming gas has to be decreased with all remaining parameters remaining constant, and this will decrease the gases velocity in the gas diffusion layer and, hence, reduce the convection heat transfer in this region. Finally, a reduced width of the land area increases the contact resistance between the bipolar plates and the gas diffusion electrodes. Since this is an ohmic loss, it is directly correlated to the land area width.

4.5. Effect of GDL thickness

The effect of gas diffusion layer thickness on the fuel cell performance is again mostly on the mass transport, as the ohmic losses of the electrons inside the gas diffusion layer are relatively small due to the high conductivity of the carbon fiber paper. A thinner gas diffusion layer increases the mass transport through it, and this leads to reduction in the mass transport loss. The molar oxygen fraction at the catalyst layer increases with decreasing gas diffusion layer thickness due to the reduced resistance to the oxygen diffusion by the thinner

![Fig. 9. Temperature distribution through the membrane for two different membrane thicknesses: (a) 0.2 mm and (b) 0.26 mm.](image)
layer. The distribution of the local current density of the cathode side depends directly on the oxygen concentration. The thicker gas diffusion layer results in more even distribution of the local current density due to more even distribution of the molar oxygen fraction at the catalyst layer. This leads to the fact that for a thinner gas diffusion layer at a constant current density, there is a much stronger distribution of current inside the cell, the maximum local current density being at the inlet under the channel area. Therefore, the maximum temperature gradient appears in the cathode side catalyst layer of the thinner gas diffusion layer case as can be seen in Fig. 8.

4.6. Effect of membrane thickness

The effect of membrane thickness on the fuel cell performance is mostly on the resistance to proton transport across the membrane. The potential loss in the membrane is due to resistance to proton transport across the membrane from the anode catalyst layer to the cathode catalyst layer. Therefore, a reduction in the membrane thickness means that the path traveled by the protons will be decreased, thereby reducing the membrane resistance and this leads to reducing the potential loss in the membrane, which, in turn, leads to less heat generation in the membrane as can be shown in Fig. 9. These results suggested

Fig. 10. Temperature distribution at the cathode side catalyst layer for two different GDL porosities: (a) 0.3 and (b) 0.5.
that reducing the membrane thickness plays a significant role in promoting cell performance.

4.7. Effect of GDL porosity

The porosity of the gas diffusion layer has two competing effects on the fuel cell performance; as the porous region provides the space for the reactants to diffuse towards the catalyst region, an increase in the porosity means that the onset of mass transport limitations occurs at higher current densities, i.e. it leads to higher limiting currents. The adverse effect of a high porosity is an expected increase in the contact resistance. Higher gas diffusion layer porosity improves the mass transport within the cell, and this leads to reducing the mass transport loss.

The molar oxygen fraction at the catalyst layer increases with more even distribution with an increase in the porosity. This is because a higher value of the porosity provides less resistance for the oxygen to reach the catalyst layer. A higher porosity evens out the local current density distribution. For a lower value of porosity, a much higher fraction of the total current is generated under the channel area. This can lead to local hot spots inside the membrane electrode assembly as can be seen in Fig. 10. These hot spots can lead to a further drying out of the membrane, thus increasing the electric resistance, which, in turn, leads to more heat generation and can lead to a failure of the membrane. Thus, it is important to keep the current density relatively even throughout the cell. As mentioned above, another loss mechanism that is important when

Fig. 11. Temperature distribution at the cathode side catalyst layer for two different values of the GDL thermal conductivity: (a) 0.5 W/m K and (b) 2.9 W/m K.
considering different gas diffusion layer porosities is the contact resistance. Contact resistance occurs at all interfaces inside the fuel cell. The magnitude of the contact resistance depends on various parameters such as the surface material and treatment and the applied stack pressure. The electrode porosity has a negative effect on electron conduction. Since the solid matrix of the gas diffusion layer provides the pathways for electron transport, the higher volume porosity increases resistance to electron transport in the gas diffusion layers.

4.8. Effect of GDL thermal conductivity

Thermal management is required to remove the heat produced by the electrochemical reaction in order to prevent drying out of the membrane and excessive thermal stresses that may result in rupture of the membrane. The small temperature differential between the fuel cell stack and the operating environment makes thermal management a challenging problem in PEM fuel cells. The temperature distribution at the cathode side catalyst layer can be shown in Fig. 11 for two cases of the gas diffusion layer thermal conductivity values. The maximum temperature with higher gradient appears in the cathode side catalyst layer of the lower thermal conductivity. Heat generated in the catalyst layer is primarily removed through the gas diffusion layer to the current collector rib by lateral conduction. This process is controlled by the gas diffusion layer thermal conductivity. Therefore, the membrane temperature is strongly influenced by the gas diffusion layer thermal conductivity, indicating a significant role played by lateral heat conduction through the gas diffusion layer in the removal of waste heat to the ambient. Therefore, a gas diffusion layer material having higher thermal conductivity is strongly recommended for fuel cells designed to operate with high power.

4.9. Optimum cell performance

In designing a cell, everything comes into play: cell potential, cell life, the pressure of the gas in the anode and cathode, operating temperature, the required mass flow of the reactants, indicating how much water is in the fuel and the oxidant, material of various elements and even the dimensions of various elements, specially the membrane, gas diffusion layers and the gas channels. Changing the cell operating parameters can have either a beneficial or a detrimental impact on fuel cell performance. The material parameters, cell design and cell operating conditions that give optimal performance depend on the application area. Stationary, portable and transportation applications all have different requirements, operate in different environments and the avail-

![Fig. 12. Optimum power with corresponding efficiency and temperature gradient for different operating conditions. Key: 1—base case, 2—cell operating temperature = 60 C, 3—cell operating temperature = 90 C, 4—cell operating pressure = 1 atm, 5—cell operating pressure = 5 atm, 6—stoichiometric flow ratio = 1.5, 7—stoichiometric flow ratio = 3, 8—GDL porosity = 0.3, 9—GDL porosity = 0.5, 10—GDL thermal conductivity = 0.5 W/m K, 11—GDL thermal conductivity = 2.9 W/m K, 12—gas channel width = 0.8 mm, 13—gas channel width = 1.2 mm, 14—GDL thickness = 0.2 mm, 15—GDL thickness = 0.3 mm, 16—membrane thickness = 0.2 mm, 17—membrane thickness = 0.26 mm.](image-url)
able fuel and oxidant conditions vary greatly. A PEM fuel cell used in a stationary application could operate from fully humidified fuel with high operating temperature and benefit from a compressor to increase air pressure, which increases the cell’s power output. In contrast, a fuel cell in portable applications such as a laptop PC or cell phone would most likely operate with air at atmospheric conditions. Furthermore, the amount of water available for fuel and air humidification in a portable fuel cell might be limited. Other requirements vary according to application. For example, fuel cell weight is much more critical in mobile appliances, and it limits the choice of materials for manufacturing. In order to determine the optimum cell performance, the cell power at various operating conditions is compared at constant nominal current density. Results with different operating conditions for the cell operated at nominal current density of 1.2 A/cm² are presented in Fig. 12. Higher operating temperature (case 3) will increase cell power and then cell efficiency and lower the cost. In addition, the maximum temperature gradient inside the cell will be reduced. Operating at a higher pressure (case 5) will increase cell power and efficiency and lower the cost. In addition, the maximum temperature gradient inside the cell will decrease. However, there will be a higher parasitic power to compress the reactants and the cell stack pressure vessel and piping will have to withstand the greater pressure, which adds extra cost. Operating at a higher stoichiometric flow ratio (case 7) will increase cell power and efficiency and lower the cost. In addition, the maximum temperature gradient inside the cell will decrease. However, there is a cost to pay for an increase in the stoichiometric flow ratio. There must be an optimum, where the gain in the cell performance just balances the additional costs of a more powerful blower. This will have to be carefully considered when designing the fuel cell system. Operating at lower or higher gas diffusion layer porosities from the base case value (cases 8 and 9) will decrease cell power and efficiency, but with higher porosity value (case 9), the maximum temperature gradient inside the cell will decrease. Operating at a higher gas diffusion layer thermal conductivity (case 11) will decrease the maximum temperature gradient inside the cell. The cell power and efficiency remain constant for both cases of higher and lower values of thermal conductivity. Operating at a narrow gas flow channel (case 12) will increase cell power and efficiency and lower the cost. In addition, the maximum temperature gradient inside the cell will decrease. Operating with a thinner membrane (case 16) will increase cell power and efficiency and lower the cost. In addition, the maximum temperature gradient inside the cell will decrease.

5. Conclusions

A full three-dimensional, multi-phase computational fluid dynamics model of a PEM fuel cell with straight flow channels has 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. This comprehensive model accounts for the major transport phenomena in a PEM fuel cell: convective and diffusive heat and mass transfer, electrode kinetics, transport and phase change mechanism of water and potential fields. The model accounts for the liquid water flux inside the gas diffusion layers by viscous and capillary forces and, hence, is capable of predicting the amount of liquid water inside the gas diffusion layers. The physics of phase change are included in this model 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 has been a function of the level of over saturation. The new feature of the algorithm developed in this work is its capability for accurate calculation of the local activation overpotentials, which, in turn, results in improved prediction of the local current density distribution. An optimization study using this model has been performed. The study quantifies the impacts of operating, design and material parameters on fuel cell performance. The model is shown to be able to: (1) understand 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 optimization of future fuel cells with much higher power density and lower cost. In addition, the results show that the model is capable of identifying important parameters for the wetting behavior of the gas diffusion layers and can be used to identify conditions that might lead to the onset of pore plugging, which has a detrimental effect on fuel cell performance, especially in the mass transport limited region.

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References


