

International Journal of Engineering Science 37 (1999) 431-452

International Journal of Engineering Science

# A two-dimensional analysis of mass transport in proton exchange membrane fuel cells

# D. Singh, D.M. Lu, N. Djilali\*

Transportation Fuelcell Systems Laboratory, Institute for Integrated Energy Systems, University of Victoria, Victoria, B.C., V8W 3P6, Canada

Received 15 January 1998

(Communicated by H. DEMIRAY)

#### Abstract

A theoretical model is developed to simulate transport phenomena in a proton exchange membrane fuel cell (PEMFC). The primary focus of this paper is the modelling and assessment of *two-dimensional* effects neglected in previous studies. The work is motivated by the need to understand the transport processes in fuel cells in order to improve heat and water management, and to alleviate mass transport limitations. The model takes into account diffusion of the humidified fuel (H<sub>2</sub>, CO<sub>2</sub> and H<sub>2</sub>O<sup>(v)</sup>) and oxidant gases (O<sub>2</sub>, N<sub>2</sub> and H<sub>2</sub>O<sup>(v)</sup>) through the porous electrodes, and convective and electro-osmotic transport of liquid water in the electrodes and the membrane. The thermodynamic equilibrium potential is calculated using the Nernst equation, and reaction kinetics are determined using the Butler–Volmer equation. A finite volume procedure is developed to solve the system of differential equations.

The model is validated against available experimental data, and numerical simulations are presented for various one- and two-dimensional isothermal cases. The results indicate that the cathode potential loss, associated with the slow  $O_2$  reaction rate, is dominant at all practical current densities. The simulations also show that two-dimensionality has a significant effect on water management and on some aspects of fuel cell performance. In particular, the anode and cathode water fluxes are found to vary considerably along the oxidant and fuel flow channels, and two new transitional water transport regimes are revealed by the two-dimensional simulations. The influences of flow configuration and electrode porosity on predicted cell performance are also discussed.  $\bigcirc$  1998 Elsevier Science Ltd. All rights reserved.

#### Nomenclature

C

concentration of a species

<sup>\*</sup> Corresponding author.

<sup>0020-7225/98/\$19.00 © 1998</sup> Elsevier Science Ltd. All rights reserved. PII: S0020-7225(98)00079-2

432	D. Singh et al.   International Journal of Engineering Science 37 (1999) 431–452
D	diffusivity of a component or a gas pair in a mixture
E	cell equilibrium potential
F	Faraday's constant, 96 487 C/mol
i	superficial current density
k	permeability
т	number of charged species
Ν	superficial molar flux of a species
n	number of species, or number of electrons participating in a reaction [Eq. (6)]
р	pressure
R	universal gas constant
Т	temperature
u	velocity
V	voltage or potential
X	mole fraction of a species
У	co-ordinate in gas supply channel direction
Ζ	charge number of a species
Ζ	co-ordinate in cell sandwich direction
Greek letters	
α	transfer coefficient
$\epsilon$	volumetric ratio of pores in a membrane or electrode
η	electrode surface potential
$\phi$	electric potential
$\mu$	viscosity
Superscripts and	d subscripts
$0^{\mathbf{d}}$	values in gas diffusers
$O^{\text{eff}}$	effective value
$0^{m}$	values in the membrane
0*	dissolved gases
$O^{0}$	reference
Oa	anode
Oact	activation
Oc	cathode
Of	ions bonded to the membrane matrix
() <sub>g</sub>	gas phase
$O_i$	species i
() <sub>ij</sub>	gas pair <i>i</i> , <i>j</i> , in a mixture
Oı	liquid water
Oohm	ohmic resistance
Op	hydraulic quantity
()s	superficial values
Ow	water
$O_z$	z component of a vector
$O_{\phi}$	electronic parameter

#### 1. Introduction

Fuel cells (FCs) are electrochemical devices that convert directly into electricity the chemical energy of reaction of a fuel (usually hydrogen) with an oxidant (usually oxygen from ambient air). A fuel cell is not a battery-like energy storage device, but rather allows continuous power generation, in the manner of a conventional engine, when supplied continuously with a fuel. Since the only by-products of the reaction in a fuel cell are heat and water, FC technology offers the prospect of zero emission energy production for applications ranging from stationary power generation for electric utilities networks to automotive transportation. Fuel cells are typically classified according to the type of electrolyte used in the cell, and several types are currently under development, ranging from high temperature solid-oxide fuel cells to low temperature proton-exchange membrane fuel cells (PEMFC). Low operating temperatures (typically in the range  $60^{\circ} \sim 90^{\circ}$ C) and a relatively simple design make PEMFCs particularly well suited to future transportation applications; their potential has, for instance, been demonstrated in the Ballard transit bus [1].

A proton-exchange membrane fuel cell, shown schematically in Fig. 1, consists of a polymer membrane sandwiched between two gas-diffusion electrodes. The electrodes are porous composites made of electronically conductive material into which the reactants (fuel and oxidant gases) diffuse and product water exits, while electrons travel through the solid portion of the electrodes. The reaction rates are enhanced by a thin catalyst layer (typically platinum) at the electrode–membrane interfaces. At the anode, fuel (H<sub>2</sub>) is oxidized liberating electrons and producing protons. The free electrons flow to the cathode via an external circuit where they combine with the protons and the dissolved oxidant  $O_2$ , to produce water (and heat). The electrolyte. The water transport through the membrane takes place via two mechanisms: electro-osmotic flow associated with the protons proceeding from the anode to the cathode, and back diffusion in the opposite direction from the high (cathode) to low (anode) concentration sites.

Fuel cells have inherently higher efficiencies than many competing energy conversion systems, but substantial progress is required to increase power density and, most importantly, reduce manufacturing cost before FCs become commercially viable. Two of the critical transport phenomena issues in PEMFC technology are: (i) thermal and water management; and (ii) mass transport limitations. Water management ensures that the polymer electrolyte membrane is maintained in the most hydrated state to ensure high conductivity and good performance. In current PEMFC designs, saturation of the membrane is maintained by humidifying the reactant gas. Thermal management is required to remove the heat produced by the electrochemical reaction (up to 50% of the energy produced during high power density operation [2]) in order to prevent drying out of the membrane and excessive operating temperature. Heat removal is a challenging problem in PEMFCs due to small differential temperature between the FC ( $80^{\circ} \sim 90^{\circ}$ C) and the operating environment, and in practice forced convection cooling is required. This entails pumping of a coolant into separate cooling channels, thus adding to the complexity and cost and reducing the system efficiency. Mass transport limitations, on the other hand, are associated with the use of air as the cathodic



Fig. 1. Operating principle of a proton exchange membrane fuel cell.

reactant, and are exhibited by a drastic degradation in performance (polarization) above a certain current density. This is attributed to a lack of oxygen reaching the catalyst, creating oxygen understoichiometry or 'starvation' at the cathode. In practice, this problem is alleviated by pressurizing the air, again at the cost of reduced system efficiency. Overall fuel cell potential losses are made up of membrane and electrodes ohmic losses, and cathode activation overpotential. With recent developments in the understanding of membrane properties and particularly with the introduction of thinner membranes and membranes of lower equivalent weight, it appears that membrane losses in well-humidified PEMFCs are not always the most important source of performance limitation. The results of Bernardi and Verbrugge [3], hereinafter BV, indicate that cathode overpotential might be the dominant component of cell potential losses.

Some fundamental aspects of heat and mass transport related to the above issues remain obscure and their elucidation has been a major challenge in PEMFC research because the transport processes involve multi-component and multi-phase flow, heat and mass transfer in porous media with electro-chemical reactions. A better understanding and proper modelling of such processes is indispensable for the improvement and optimization of the design of FC systems, and this has prompted the formulation of a number of models in recent years. Due to the complexity of the processes and the difficulty in obtaining closed form solutions, onedimensional isothermal conditions are assumed in most models proposed in the literature. The models of Verbrugge and Hill [4], Springer et al. [5], Bernardi and Verbrugge [3, 6], and Fuller and Newman [7] all use the Stephan-Maxwell equation for gas transport and introduce additional equations to account for mass transport limitations. All these models have provided useful insight and reasonable predictions of the cell performance in the low and intermediate current density ranges, but fail to reproduce the abrupt drop observed experimentally at higher current densities. Springer et al. [5] introduced in their model an expression to account for the porosity and tortuosity of the electrode and showed much improved agreement in the mass transport limited range. More recently, Mosdale and Srinivasan [2] showed that a simplistic approach focussing on mass transport limitations in the catalyst layer can lead to a good fit with experimental data. A two-dimensional (2-D) isothermal model was presented by Nguyen and White [8], who considered transport is in the membrane only and used an empirical correlation to account for the electro-osmotic drag.

One-dimensional isothermal models have provided useful insight and fundamental understanding, but remain applicable only to single cells with relatively simple geometries and flow channel configurations. In stack modelling, multi-dimensional diffusive and convective heat and mass transfer have to be taken into account in the membrane, electrodes and gas channels if reliable simulation and design tools are required over a range of operating conditions and scales. The development of such a general model remains a very challenging task from the viewpoints of modelling as well as numerical analysis. However much progress has been made in recent years in the development of powerful and accurate algorithms, and it is now possible to solve the full set of multi-dimensional transport equations of fluid flow, heat and mass transfer for many complex situations. As a stepping stone towards a general model, we present in this paper a 2-D isothermal analysis for a complete fuel cell, including the anode diffusion layer neglected in previous studies. The model accounts for: (i) electron transport; (ii) diffusion of gases in both anode and cathode; (iii) electrochemical reaction activation; (iv) migration of protons from anode to cathode via the membrane; and (iv) transport of water through the membrane and the electrodes.

## 2. Mathematical model and numerical solution

The model is formulated for a single PEMFC. A schematic of the cell and the coordinates system are shown in Fig. 2.

## 2.1. Model assumptions

In this study, the fuel cell is assumed to operate under steady and uniform temperature conditions. Gases are assumed perfect, and the variation of total gas pressure within the gas



Fig. 2. Schematic view of the elements of a PEMFC and co-ordinate system used in the model.

diffuser portion of the electrodes is considered negligible compared with the variation of the hydraulic pressure, since the gas-phase is much less viscous than the liquid-phase. Gases in the electrodes are assumed to be saturated with moisture. The catalyst layers are much thinner than either the electrode diffuser or membrane, and are treated as thin interfaces.

The following transport phenomena are taken into account in the model:

- 1. transport of electrons through the carbon electrodes;
- 2. multi-component diffusion of gaseous species through the porous anode and cathode;
- 3. flow of liquid and vapour water through anode and cathode;
- 4. proton migration through the membrane;
- 5. transport of water through the membrane via diffusion and electro-osmotic convection;
- 6. electrochemical reaction at the catalyst.

#### 2.2. Governing equations

The basic equations used in the model are given below, and a more detailed description of the mathematical model is deferred to the Appendix. Diffusion of the multi-component gas streams through the electrodes may be described using the Stefan–Maxwell equations [10], i.e.

$$\nabla x_i = \sum_{j=1}^n \frac{RT}{p_{\rm g} D_{ij}^{\rm eff}} (x_i \mathbf{N}_{j\rm g} - x_j \mathbf{N}_{i\rm g}), \quad i = 1, 2, \dots, n-1,$$
(1)

where the various symbols are described in the Nomenclature, and bold characters denote vectors.

437

The liquid water transport in the electrodes is governed by Darcy's law

$$\mathbf{u}_{\mathrm{l}}^{\mathrm{d}} = -\frac{k_{\mathrm{p,s}}^{\mathrm{d}}}{\mu_{\mathrm{l}}} \nabla p_{\mathrm{l}},\tag{2}$$

whereas liquid water flow in the membrane is modelled by the Schlögl equation, in which an additional term is introduced to account for the electro-osmotic drag [11]

$$\mathbf{u}_{l}^{m} = \frac{k_{\phi}}{\mu_{l}} Z_{f} C_{f} F \nabla \phi - \frac{k_{p}^{m}}{\mu_{l}} \nabla p_{l}.$$
(3)

The flux of protons migrating through the membrane is given by the Nernst–Planck equation [12]

$$\mathbf{N}_{i} = -Z_{i} \frac{F}{RT} D_{i} C_{i} \nabla \phi - D_{i} \nabla C_{i} + C_{i} \mathbf{u}_{1}^{\mathrm{m}}, \quad i = 1, 2, \dots, m,$$

$$\tag{4}$$

where the three terms on the right-hand side represent the fluxes due to electric field, concentration gradient and convective transport, respectively. The flow of charged species in the membrane is related to the electric current via

$$\mathbf{i} = F \sum_{i=1}^{m} Z_i \mathbf{N}_i.$$
(5)

The electrochemical reaction is modelled using first-order reaction kinetics as described by the Butler–Volmer equation [13]

$$|\mathbf{i}| = i_0 [\mathrm{e}^{-\alpha n F \eta/RT} - \mathrm{e}^{(1-\alpha) n F \eta/RT}], \tag{6}$$

where  $i_0$  is the empirically determined exchange current density [3].

Mass conservation for species and current continuity, together with auxiliary equations (equation of state for gases, equilibrium potential of a cell) are combined with Eqs. (1)–(6) to obtain the complete set of model equations describing transport processes in a PEM fuel cell. The equations and associated boundary conditions are given in the Appendix. These equations are solved numerically for  $x_i$ ,  $p_i$ ,  $\phi$ ,  $N_i$ ,  $u_i$ , i.

#### 2.3. Numerical solution procedure

A finite volume method is used to solve the differential equations of the 2-D model. A staggered grid system is introduced in order to eliminate numerical 'checker board' oscillations encountered with the single grid system. In this arrangement species concentration, hydraulic pressure and electric potential are discretized and computed on the main grid, whereas fluxes, liquid water velocity and electric current are computed at the staggered nodes located mid-way between the main grid nodes. The algebraic sets of discretized equations are solved using a block iterative technique with under-relaxation.

The influence of grid resolution on the accuracy of numerical solution was investigated over a range of cell operating conditions using various numerical grid spacings. A grid with



Fig. 3. Comparison of predicted performance (polarization) curve with experimental data.

 $54 \times 108$  (in  $z \times y$ ) nodes was found to provide adequate resolution, yielding computed current densities within 0.5% of the values obtained with finer grids. This grid density is used for all the simulations presented in this paper.

## 3. Results and discussion

The numerical simulations presented in this paper focus mainly on 2-D effects on PEMFC performance. The physical properties of the membrane and electrode are the same as those given in BV [3].

## 3.1. Model validation

To validate the computer code and to allow comparison with the 1-D results of BV [3], the model was first used as a pseudo 1-D model by assuming constant concentration and corresponding fluxes on the electrode-channel boundaries. Fig. 3 compares the i-V curves computed with the two models to the measurements of Ticianelli et al. [9]. The results of the



Fig. 4. Computed variation of hydraulic pressure: 1-D simulation;  $i = 105 \text{ mA/cm}^2$ .

present model are in very good overall agreement with both those of BV [3] and experimental data. It should be pointed out that in their model, Bernardi and Verbrugge did not account for the anode gas diffusion and did not treat the catalyst layers as interfaces; this might account for the slight difference between the two models. Fig. 4 shows the distribution of hydraulic pressure in the fuel cell calculated by the present model under the 1-D assumption. The results are consistent with those of BV (cf. Fig. 8 of ref. [3]).

## 3.2. 2-D effects

In the 1-D case, the concentration of the reactants in the gas supply channels are assumed to remain constant. In actual operation, the concentrations drop along the channels as the reactants are consumed. The determination of the mass transfer coefficients along the gas channel–electrode interfaces would require the computation of convective mass transfer in the gas channels, which is beyond the scope of this initial study. Instead, the two-dimensional effects are introduced by allowing concentrations to vary linearly along the gas channels. This is a simplified boundary condition which does not account for non-linear effects, yet it is much more realistic than the 1-D condition (constant concentration) assumed in previous studies.

Typical computed distributions of membrane electric potential, current density, hydraulic pressure and liquid water velocity shown in Fig. 5–8 clearly illustrate the 2-D nature of the transport processes. A comparison of cell performance, i.e. i-V curve, obtained from the 2-D and 1-D simulations under the same base case conditions (see Table 1) is presented in Fig. 9.



Fig. 5. 2-D distribution of electric potential in the membrane;  $i_{av} = 56 \text{ mA/cm}^2$ .



Fig. 6. 2-D distribution of electric current;  $i_{av} = 56 \text{ mA/cm}^2$ .



Fig. 7. 2-D distribution of hydraulic pressure;  $i_{av} = 56 \text{ mA/cm}^2$ .

The cell voltage predicted for the 2-D case is lower than the 1-D estimate, particularly at low current densities. We note that the voltage and current density for the 2-D case are averaged over the channel length.

Fig. 10 shows the various contributions to cell potential loss. The cathode overpotential appears to be the dominant factor in cell performance, accounting for 60-70% of the total cell loss, with the membrane resistance emerging as the second largest contributor. While, anode overpotential is negligible for most practical current densities, the trend indicates that it might become significant at very high current densities.

## 3.3. Effect of flow configurations on cell performance

The effect of gas channel flow configurations such as counter-flow (i.e. fuel and oxidant gases flowing in opposite directions in the anode and cathode side supply channels) and coflow arrangements can be investigated with the 2-D model. The cell performance curves for the co- and counter-flow cases, shown in Fig. 11, show that the two flow arrangements result in nearly the same output voltage until i = 1.23 A/cm<sup>2</sup>, at which point severe polarization takes place in the counter-flow case and the cell voltage drops abruptly as a result of reactant deficiency or mass transfer limitations. In the co-flow case the corresponding voltage drop does not take place till a much higher current density ( $i \approx 2.4$  A/cm<sup>2</sup>). Use of higher oxidant



Fig. 8. 2-D variation of liquid water velocity in the membrane and electrodes;  $i_{av} = 56 \text{ mA/cm}^2$ .

 Table 1

 Base case operating conditions and physical parameters

Quantity	Base-case conditions value	
Wet membrane thickness	0.023 cm	
Gas diffusion electrode thickness	0.026 cm	
Gas diffusion electrode porosity	0.40	
Relative humidity of fuel	100%	
Relative humidity of air	100%	
Cell temperature	95°C	
Anode stoichiometric flow ratio $\psi_a$	1.3	
Cathode stoichiometric flow ratio $\psi_c$	3.0	
$x_{N_2}/x_{O_2}$	0.79/0.21	
X <sub>CO2</sub>	5 ppm	
Anode chamber pressure $P_{\rm a}$	3.0 atm	
Cathode chamber pressure $P_{\rm c}$	5.0 atm	



Fig. 9. Variation of cell potential vs current: comparison of 1-D and 2-D simulations.

concentration and/or higher stoichiometric flow ratio would shift the limiting current to higher values.

## 3.4. Effect of electrode porosity on cell performance

Electrode porosity has a significant influence on the transport of reactant gases, since cell performance may be seriously hampered by an insufficient supply of reactants through the electrode diffusers. Fig. 12 demonstrates the adverse effect of low porosity on cell performance. For  $\epsilon = 0.11$ , the cell voltage drops rapidly at  $i \sim 0.22$  A/cm<sup>2</sup>, due to the mass transfer limitation, whereas the corresponding limiting current density is greater than 3.0 A/cm<sup>2</sup> for a porosity of  $\epsilon = 0.5$ .

#### 3.5. 2-D effect on water management

Water management is of crucial importance to achieving satisfactory cell performance and high power densities. Due to the electro-osmotic effect and the heat generated by the oxygen reduction reaction at the cathode catalyst layer, the membrane tends to dehydrate; this results



Fig. 10. Contributions of membrane, anode and cathode overpotential to overall fuel cell losses.

in increased membrane resistance and, consequently, loss of cell potential. The tendency for dehydration is exacerbated at higher current densities. In addition, at high current densities more water is produced at the cathode. If the excess water is not removed effectively, flooding of the electrode occurs resulting in a lack of reactant reaching the catalyst layer and creating an understoichiometry of the reactant gases which leads to severe concentration polarization.

The two-dimensional water transport discussed earlier in the context of Fig. 8, indicates that the membrane may experience various levels of dehydration along the gas channels. Water management requirements along both anode and cathode gas channels vary significantly depending on operating conditions, and the 2-D simulations reveal five regimes:

- 1. At very low current densities (e.g.  $i_{av} < 50 \text{ mA/cm}^2$ , for base case conditions), 'back diffusion' takes place, i.e. liquid water flows from cathode to anode along the entire channel length as shown in Fig. 13(a). This indicates that additional liquid water is required at the entire cathode–channel interface, while there are no additional humidification requirements on the anode side. Due to the variation of water fluxes along the channel, higher humidification levels may be desirable downstream of the inlet.
- 2. At low current densities, liquid water flows out of both anode and cathode near the inlet region of the channels, whereas further downstream, the pattern reverts to a similar one to



Fig. 11. Comparison of predicted cell performance for co- and counter-flow cases.

that of regime 1) above. Hence there is no additional humidification requirement near the inlet of the electrode-channel boundaries, but humidification may be needed further downstream [see Figs. 8 and 13(b)].

- 3. At intermediate current densities (up to  $\approx i_{av} = 200 \text{ mA/cm}^2$ , for base case conditions), liquid water flows out of both sides of the fuel cell throughout the channel length [Fig. 13(c)]. The water produced by the O<sub>2</sub>, reduction reaction is partitioned. Hence, no additional water is required on either side of the cell.
- 4. At high current densities (up to  $\approx i_{av} = 2000 \text{ mA/cm}^2$ , base case conditions), liquid water flows from anode to cathode near the inlet of the electrode-channel interface, and further downstream the pattern changes to one similar to regime (3) as illustrated in Fig. 13(d). Additional water is required in the neighbourhood of the inlet region of the anode side (positive flux), while no humidification is required further downstream. Computed liquid water velocity distributions at a high current density corresponding to this regime are shown in Fig. 14.
- 5. At very high current densities, liquid water flows from anode to cathode along the entire channel–electrode interface as shown in Fig. 13(e), and additional humidification is required along the entire anode side to prevent membrane dehydration.



Fig. 12. Effect of electrode porosity on cell performance (co-flow case).

BVs study [3] showed the existence of regimes (1), (3) and (5) only, with clear-cut boundaries between the three regimes as a result of the 1-D assumption. The two new transitional regimes (2) and (4) revealed by the present 2-D simulations are accompanied with much narrower current density ranges for regimes (1) and (3) than predicted by the 1-D model. We note that the above results are obtained for cases where a back pressure is applied to the gas supply channels, i.e.  $p_c > p_a$ , the usual operating condition of PEMFCs. Under balanced pressure conditions, the current density ranges for the various regimes would be different.

In the new transitional regimes, more elaborate water management schemes may be beneficial to cater for the fact that only part of an electrode may need to be humidified, while the product water in the remainder of the electrode can be removed by the inherent water transport processes. If an electrode is humidified along its entire length regardless of the water flow pattern, local flooding may occur. In regime (2), for instance, humidification is only required further downstream in the cathode gas supply channel instead of near the inlet, as illustrated in Fig. 13. Conversely, in regime (4), humidification requirements, as deduced from Fig. 14, are primarily near the inlet of the anode side (positive flux) and vanish almost immediately downstream of the inlet. At still higher current densities, in regime (5), humidification requirements are significant near the inlet and drop rapidly downstream, and a



(5)

Fig. 13. Water transport regimes at various current densities: (a)  $i_{av} \leq 50 \text{ mA/cm}^2$ ; (b)  $50 \text{ mA/cm}^2 \leq i_{av} \leq 150 \text{ mA/cm}^2$ ; (c)  $150 \text{ mA/cm}^2 \leq i_{av} \leq 200 \text{ mA/cm}^2$ ; (d)  $200 \text{ mA/cm}^2 \leq i_{av} \leq 2000 \text{ mA/cm}^2$ ; (e)  $i_{av} \geq 2000 \text{ mA/cm}^2$ . (Co-flow case with gas flow direction from bottom to top; boundaries between regimes correspond to base conditions in Table 1.)



Fig. 14. Variation of liquid water velocity in the membrane and electrodes: co-flow case;  $i_{av} = 220 \text{ mA/cm}^2$ .

water management scheme whereby humidification is reduced further downstream of the anode channel might be effective in preventing premature flooding of the anode near the outlet.

## 4. Conclusions

A 2-D numerical analysis of transport processes in a proton-exchange membrane fuel cell is presented. Two-dimensional effects, previously neglected, are found to be important and to have a significant impact on some aspects of FC operation and water management in particular. The FC performances predicted by the 2-D model are, in general, more conservative than those obtained assuming 1-D processes. Specifically, the 2-D model results in: (i) slightly lower cell voltage; (ii) severe concentration polarization taking place at much lower current densities; and (iii) increased humidification requirements at lower current densities. Cathode overpotential, associated with the slow  $O_2$  reaction rate, is found to be the dominant contribution to cell potential loss dominant at all practical current densities.

The 2-D simulations reveal two new regimes in which it could be beneficial to implement more complex water management schemes, whereby only the electrode would be humidified only along a portion of the flow channel, while product water in the remainder would be removed by water transport processes inherent to the operation of a fuel cell. Another interesting 2-D feature of the present simulations is the abrupt concentration polarization which is predicted to occur at lower current densities for the counter-flow case, indicating that the co-flow arrangement mitigates mass transport limitations.

In this study, temperature non-uniformity has not been accounted for. In many cell and stack configurations non-isothermal effects can become important, particularly at high current densities, and a coupled heat and mass transport analysis is then required to investigate heat and water management issues and to predict cell performance. The extension of the present 2-D model to non-isothermal cases is currently under way. Furthermore, linear variations of the gas concentrations were assumed along the electrode-flow channel interfaces in order to make the problem numerically tractable. An approach that couples the present model with a computational model of the convective mass transport in the flow channels would eventually be required to fully account for and assess the effect of non-linear and entrance effects.

## Acknowledgements

This research was performed under a Collaborative Research and Development (CRD) Grant funded by British Gas Investments (Canada), Ballard Power Systems Inc. and the Natural Sciences and Engineering Research Council of Canada (NSERC).

## Appendix A

## A.1. Complete mathematical model and boundary conditions

The anode and cathode gas streams consist of three gas species each:  $i = (H_2, CO_2, H_2O^{(v)})$ , or  $i = (O_2, N_2, H_2O^{(v)})$ , respectively. The complete set of equations describing the transport processes in the electrodes and membrane are given below.

A.1.1. Electrodes

$$\nabla x_i = \sum_{j=1}^{3} \frac{RT}{p_{\rm g} D_{ij}^{\rm eff}} (x_i \mathbf{N}_{j\rm g} - x_j \mathbf{N}_{i\rm g}), \quad i = 1, 2$$
(A1)

 $\nabla \cdot \mathbf{N}_{ig} = 0, \quad i = 1, 2, 3 \tag{A2}$ 

$$\sum_{i=1}^{3} x_i = 1 \tag{A3}$$

$$x_i = p_{ig}/p_g \tag{A4}$$

$$x_{3 g} = xwg = constant < q > (A5)$$

$$\nabla^2 p_1 = 0 \tag{A6}$$

$$\mathbf{u}_{l}^{d} = -\frac{k_{p,s}^{d}}{\mu_{l}} \nabla p_{l}.$$
(A7)

## A.1.2. Membrane

$$\nabla^2 p_1 = 0 \tag{A8}$$

$$\nabla^2 \phi = 0 \tag{A9}$$

$$\mathbf{u}_{l}^{m} = \frac{k_{\phi}}{\mu_{l}} Z_{f} C_{f} F \nabla \phi - \frac{k_{p}^{m}}{\mu_{l} \nabla p_{l}}$$
(A10)

$$\mathbf{i} = -\kappa \nabla \phi + F C_{\mathrm{f}} \mathbf{u}_{\mathrm{l}}^{\mathrm{m}}.$$
(A11)

Equations (A8)–(A11) are obtained by applying the principles of mass and current continuities and electro-neutrality. The protonic conductivity,  $\kappa$  in eqn (A11) is expressed as:

$$\kappa = F^2 D_{\rm H} + C_{\rm f}/RT \tag{A12}$$

The total cell voltage is calculated as

$$V = E - \eta_{\rm act} - \Delta V^{\rm m} - \Delta V^{\rm d}_{\rm ohm},\tag{A13}$$

where the equilibrium potential E is evaluated from the Nernst equation, which provides a relationship between E and  $E^0$  the reversible cell potential at standard temperature

$$E = E^{0} + \frac{RT}{nF} \ln(C *_{H_{2}} C *_{O_{2}})$$
(A14)

 $\eta_{act}$  the activation polarization, is obtained using Eq. (6), and

$$\Delta V^{\rm m} = \Delta \phi^{\rm m}. \tag{A15}$$

# Boundary conditions

Boundary conditions are required along the fuel channel-anode interface (z = 0), the air channel-cathode interface  $z = z_c$ , the catalyst interfaces  $(z = z_{am} \text{ and } z = z_{cm})$ , and along the side boundaries of the cell (y = 0 and y = L). These boundary conditions are:

450

at z = 0

- mole fraction: linear profiles are assumed for  $x_i$ , i = 1, 2, 3, and species fluxes along the interface are obtained from mass conservation.
- pressure:  $p_1 = p_a$ ;

at  $z = z_{am}$ , material balance requires

- $(C_{l}u_{l,z}^{d} + N_{wg,z})|_{z=z_{am}^{-}} = C_{l}u_{l,z}^{m}\epsilon_{w}^{m}|_{z=z_{am}^{+}};$
- $\phi = 0;$
- $i_z |_{z=z_{am}^+} = i_z |_{z=z_{am}^-};$
- at  $z = z_{cm}$ , again from material balance considerations
- $(-C_{l}u_{l,z}^{d} + N_{wg,z})|_{z=z_{am}^{+}} + \frac{i_{z}}{2F}|_{z=z_{cm}} = C_{l}u_{l,z}^{m}\epsilon_{w}^{m}|_{z=z_{cm}^{-}};$

at  $z = z_c$ 

- mole fraction: linear profiles are assumed for  $x_i$ , i = 1, 2, 3, and species fluxes along the interface are obtained from mass conservation;
- pressure:  $p_1 = p_c$ ;
- at y = 0, and y = L
- a zero flux condition  $\partial()/\partial y = 0$  is assumed for the variables  $x_i$  (i = 1, 2, 3),  $p_1$ , and  $\phi$ .

## References

[1] K.B. Prater, Solid polymer fuel cell developments at Ballard, Journal of Power Sources 37 (1992) 181–188.

- [2] R. Mosdale, S. Srinivasan, Analysis of performance and of water management in proton exchange membrane fuel cells, Electrochimica Acta 40 (4) (1995) 413–421.
- [3] D.M. Bernardi, M.W. Verbrugge, A mathematical model of the solid-polymer-electrolyte fuel cell, Journal of the Electrochemical Society 139 (9) (1992) 2477–2491.
- [4] M.W. Verbrugge, R.F. Hill, Transport phenomena in perfluorosulfonic acid membranes during the passage of current, Journal of the Electrochemical Society 137 (4) (1990) 1131–1138.
- [5] T.E. Springer, T.A. Zawodzinski, S. Gottesfeld, Polymer electrolyte fuel cell model, Journal of the Electrochemical Society 138 (8) (1991) 2334–2342.
- [6] D.M. Bernardi, M.W. Verbrugge, Mathematical model of a gas diffusion electrode bonded to a polymer electrolyte, AIChE Journal 37 (8) (1991) 1151–1163.
- [7] T.F. Fuller, J. Newman, Water and thermal management in solid-polymer-electrolyte fuel cells, Journal of the Electrochemical Society 140 (5) (1993) 1218–1225.
- [8] T.V. Nguyen, R.E. White, A water and heat management model for proton-exchange-membrane fuel cells, Journal of the Electrochemical Society 140 (8) (1993) 2178–2186.
- [9] E.A. Ticianelli, C.R. Derouin, A. Redondo, S. Srinivasan, Methods to advance technology of proton exchange membrane fuel cells, Journal of the Electrochemical Society 135 (9) (1988) 2209–2214.
- [10] R.B. Bird, W.E. Stewart, E.N. Lightfoot, Transport Phenomena. Wiley, New York, 1960.
- [11] R. Schlögl, Zur Theorie der Anomalen Osmose, Zeitschrift Physik Chemie 3 (1955) 73.
- [12] A.J. Appleby, Characteristics of fuel cell systems. in: L. Blomen, M. Mugerwa (Eds.), Fuel Cell Systems, Plenum, New York, 1993, pp. 157–199.
- [13] J. Newman, Electrochemical Systems, Prentice-Hall, Englewood Cliffs, NJ, 1973.