Computational modelling of polymer electrolyte membrane (PEM) fuel cells: Challenges and opportunities

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Abstract

Fuel cells are still undergoing intense development, and the combination of new and optimized materials, improved product development, novel architectures, more efficient transport processes, and design optimization and integration are expected to lead to major gains in performance, efficiency, reliability, manufacturability and cost-effectiveness. Computational fuel cell engineering (CFCE) tools that allow systematic simulation, design and optimization of fuel cell systems would facilitate the integration of such advances, allow less heavy reliance on hardware prototyping, and reduce development cycles.

CFCE requires the robust integration of models representing a variety of complex multi-physics transport processes characterized by a broad spectrum of length and time scales. These processes include a fascinating, but not always well understood, array of phenomena involving fluidic, ionic, electronic and thermal transport in concert with electrochemical reactions. In this paper, we report on some progress in both fundamental modelling of these phenomena, as well as in the development of integrated, computational fluid dynamics (CFD) based models for polymer electrolyte membrane (PEM) fuel cells. A new rational model for coupled protonic and water transport in PEMs, as well direct numerical simulations of two-phase flow in porous gas diffusion electrodes are discussed. Illustrative applications of CFD-based simulations are presented for conventional fuel cells and novel micro-structured fuel cells. The paper concludes with a perspective on some of the remaining theoretical, experimental and numerical challenges to achieve truly functional CFCE tools.

Keywords: Fuel cell; Transport phenomena; Two phase flow; Modelling; CFD; Polymer electrolyte membranes; Porous electrodes; Computational design; Micro-fuel cell; Review

1. Introduction

Fuel cells and hydrogen technologies offer a promising pathway towards sustainable energy systems. They offer the prospect of low to zero emission power generation for subWatt to MWatt applications in transportation, heating, manufacturing, and communication. However, many engineering and scientific obstacles (as well as economic and socio-political ones) need to be surmounted before wide spread adoption of these technologies becomes possible.

The potential of fuel cells for clean and efficient energy conversion have long been recognized. Fuel cells are adaptable, have a high theoretical efficiency, and, unlike batteries, they can produce electricity continuously as long as supplied with a fuel. When fuelled by hydrogen, the only by-products of a fuel cell are water and heat. In addition to the important question surrounding the development of a hydrogen fuelling infrastructure discussed elsewhere [1], the main obstacles to the deployment and commercialization of polymer electrolyte membrane fuel cells (PEMFCs) are cost, durability, and performance [2]. In this paper we will focus on the role of computational tools in addressing some of the technical challenges in the development of better performance and affordable PEMFC technology, the lead contender for replacing the ubiquitous battery and internal combustion engine.

The overall operating principle of a PEMFC is relatively simple. Hydrogen is fed to the cell and oxidized at the anode, while oxygen, usually carried in an air feed stream, is reduced at the cathode. The protons produced at the anode are conducted through the polymer electrolyte...
membrane (PEM) to the cathode. Since the membrane is not electronically conductive, the electrons released at the anode have to flow along an external circuit, thereby generating useful electric power, and eventually recombine with the protons on the cathode side to produce water. These reactions and pathways are illustrated schematically in Fig. 1.

Broadly speaking, improved performance in PEMFC can be achieved by enhancing the rates of transport and/or reaction, and by increasing the surface/volume ratio of interfacial areas where reactions/transport take place. This can be achieved by, e.g., engineering the transport properties or processes in specific areas, or by introducing new materials or designs. A particularly promising avenue is the use of micro-structured features [3] as will be discussed later. In any case, it is first necessary to identify the prevailing limitations and their location, and to understand their origin. In-situ measurements to obtain the relevant information are exceedingly difficult due to the confined and electrochemically active environment of fuel cells, the complexity of the couplings, and the often transient nature of the processes. Rational models based on the fundamental physico-chemical processes together with experimental observation are thus an essential tool to gain a more complete understanding of the operation of a fuel cell. Coupling these models with modern computational fluid dynamics (CFD) and multi-disciplinary optimization methods would allow the creation of powerful computational fuel cell engineering (CFCE) tools that could lead to dramatic reductions in lead times and development costs, and spur innovative design.

While much progress has been made in recent years in CFD based modelling of fuel cells, truly functional and predictive capabilities remain a challenge due to, amongst others factors:

- lack of sufficiently general models for ionic and water transport in polymer membranes;
- deficiencies in models for two phase transport in porous gas diffusion electrodes and gas flow channels;
- inadequate macroscopic modelling and resolution of catalyst layers;
- limited data for material and transport parameters and lack of in situ data for validation; and
- existence of a wide range of characteristic length and time scales.

An excellent overview of the potential gains to be made by theoretical modelling was recently provided in [4], and many notable advances in CFD modelling of PEM fuel cells have been made by several groups over the last few years [5–9], and a thorough review was provided in 2004 in [10]. Some representative contributions made since that review include parallelized three-dimensional CFD modelling [11–13]; parametric computational studies in conjunction with experimental validation [14,15]; and developments addressing the important aspects of two-phase transport [16,17] and catalyst layer modelling [18,19].
A review of challenges and modelling issues in both low and high temperature fuel cells, with particular attention to heat management, appeared recently [20]. In this paper, we provide an overview focused primarily on recent progress made at the University of Victoria in addressing some of the key transport phenomena issues and in the development of CFD based modelling strategies for PEMFCs. The first part of the paper focuses on fundamental modelling of specific transport phenomena encountered in component layers of the membrane electrode assembly (MEA). In the second part, we discuss the integration of some of these sub-models into multi-dimensional CFD codes and illustrate their application in plate and frame unit cells as well as in novel micro-structured designs. The paper closes with a perspective on some of the remaining challenges.

2. Transport phenomena in PEMFCs

The cathode side of the MEA, shown schematically in Fig. 2, provides a good illustration of the complex interplay between various transport phenomena. The MEA consists of a proton exchange membrane sandwiched between catalyst and gas diffusion layers (GDL), with the latter two components essentially forming the electrode. This electrode is a buffer zone that facilitates a number of processes:

- The conduction of electrons between the current collectors and the reaction sites.
- The conduction of protons between the membrane and catalyst layer.
- The transport of the reactants to the catalyst layer.
- The transport of heat away from the reaction site primarily through the solid matrix.
- Condensation/evaporation and transport of liquid water and vapour.

Optimal tailoring of an effective electrode requires balancing of often conflicting requirements. On the cathode side for instance, the reaction is prone to mass transport limitations at high current densities, when depletion of oxygen in the air stream becomes significant. This can be further compounded by flooding of the electrode pores by the product water and blocking of pathways for oxygen transport to the reaction sites. Effective removal of excess water is thus essential, but must be balanced against the need to prevent drying of the adjacent proton conducting media (membrane), which not only causes deterioration in protonic conductivity (and thus higher ohmic losses and lower efficiency), but also impacts durability.

Computational design of fuel cells with effective reactant distribution, optimized structures and proper heat and water management, requires understanding of the various transport processes in each of the components, the development of physically representative models for these processes, and, finally, proper coupling of the models.

2.1. Polymer electrolyte membranes

Polymer electrolyte membranes (PEM) provide the key functions of conducting protons from anode to cathode, and of preventing reactant crossover. These membranes consist of microphase-separated structures, comprising hydrophobic polymer chains (PTFE/Teflon) and hydrophilic sulfonic acid groups. A thorough review of transport phenomena in PEM was recently provided in [21]. The protonic conductivity characteristics of a PEM are strongly dependent on the membrane hydration given in terms of \( \lambda \) (number of sorbed waters per sulfonate head). Excess protons in bulk water are primarily in the form of hydronium ions (H\(_3\)O\(^+\)), and higher complexes such as “Eigen” ion (H\(_9\)O\(_4\)\(^+\)) may also participate. A phenomenon closely linked to membrane conductivity is electro-osmotic drag (EOD), the process whereby a given number of water molecules associated with a proton are dragged as the proton migrates from anode to cathode. This EOD mechanism plays a major role in determining overall water balance.

The breadth of PEM transport models proposed to date ranges from molecular dynamics simulations and statistical mechanics models, to empirical macroscopic models as illustrated in Fig. 3. Our focus is on the development of macroscopic models required for computational simulations of complete fuel cells. The most widely used model to date for determining membrane conductivity in large scale fuel cell models is due to Springer, Zawodzinski and Gottesfeld [22]. This “diffusion” model has limited predictive abilities as it relies on empirical correlations.
obtained for Nafion membranes only [23,24], and in particular does not perform well under low humidification conditions [23].

A more fundamental and general approach to macroscopic modelling transport in membranes requires microscopic consideration of the dominant interactions between the aqueous system and the polymer and how they affect the conductivity and transport properties. A general model that considers such interactions based on the fundamentals of multi-component diffusion was recently derived. This model proceeds from a generalization of the Stefan–Maxwell relations to a free solution in a membrane pore structure [25]. Specializing this formulation to polymer electrolyte membranes (specifically perfluorosulfonic acid membranes) and expressing the electrochemical potential gradient in terms of composition, activity, Gibb’s free energy, and electrical potential yields a general expression that can be simplified through an order of magnitude analysis [26]. Considering a limiting case scenario with a pressure drop of order \((5 \times 10^{5} \text{N/m}^2)\) and a potential drop of order \((0.3 \text{V})\), shows that the dominant driving terms are the potential and mole fraction gradients, \(r_{Fm}\) and \(r_{X}\).

The resulting simplification and expressing \(X\) in terms of membrane water content \(\lambda\) yields the new Binary Friction Membrane Model (BFM2) [26]:

\[
\begin{bmatrix}
N_p \\
N_w 
\end{bmatrix} = -c \begin{bmatrix}
D_{pp} & D_{pw} \\
D_{wp} & D_{ww} 
\end{bmatrix} \begin{bmatrix}
\nabla \Phi_m \\
\nabla \lambda
\end{bmatrix}.
\]

The BFM2 provides a general constitutive relation coupling the protonic flux \(N_p\), which is proportional the ionic current \(i_m\) to the water flux \(N_w\); \(c\) is the molar concentration of water; \(f = F/(RT)\), and \(\Phi_m\) and \(\lambda\) are the membrane potential and water content. The coupling between the protonic and water transport is through the diagonal transport parameters \(D_{pp}\) and \(D_{ww}\) which are considered to be functions of water content \(\lambda\) and temperature \(T\). A feature of the model is that all parameters can be obtained using a simplified version of the model obtained in the limit of conditions representing AC impedance conductivity measurements. The BFM2 has a number of features that make it promising. The model:

- relies on rationally derived transport equations based on the physics of multi-component transport in the membrane;
- is not restricted by the assumption of equimolar counter diffusion;
- accounts for the effect of temperature on the sorption isotherm;
- is not restricted to Nafion, but applies to the entire family of perfluoro-sulfonated membrane.

Fig. 4 illustrates the enhanced capabilities of the BFM2 model in capturing the variation of the conductivity for all water contents. Fig. 5 illustrates the predicted water crossover coefficient \(WC = N_{i}/[I/(2F)]\). In the second part of this paper, the BFM2 is coupled to a fuel cell model and we will see that the resulting overpotentials and overall performance predicted by the BFM2 can, for some operating conditions, differ significantly from those obtained using the classical Springer model.

2.2. Gas diffusion electrodes

The gas diffusion layers (GDLs) consist of an anisotropic fibrous structure, either in the form of a “paper” or a “woven cloth”, that allows distribution of reactant gases through the porous matrix and collection of current through the fibres (see Fig. 6). At higher current densities, or when local water and heat management is inadequate, water condensation takes place leading to potential “flooding” within the GDL (and catalyst layer) as discussed earlier. In order to remedy this, GDLs are commonly treated with PTFE (Teflon) to impart hydrophobicity, and force water droplets to agglomerate at the free surface of the GDL. However, the hydrophilic Nafion in the catalyst layer will absorb and retain liquid water.

Fig. 4. Calculated conductivity using BFM2 (solid) and models of Springer et al. [22] and Thampan et al. [24] against experimental data [27] for E-form Nafion 117 (dashed bounds).
Thus, liquid water produced is transported from a saturated catalyst layer to the free surface of the GDL.

A major stumbling block in the reliability of two-phase models that attempt to account for liquid water transport in GDLs is the necessity to rely on empirical correlations and experimental observations obtained in soil and sand samples—media with a structure that differs significantly from fibrous or woven GDLs.

The phase distribution in a porous media is potentially the result of viscous, capillary, and gravitational forces. When modelling the diffusion layer of a PEMFC, the second (liquid) phase is taken to consist of a single component, with transfer of water only across the phase boundary through evaporation/condensation. For porous media in which the void space is occupied by two-phases, the bulk porosity \( e \) is divided between the liquid \( e_1 \) and gas \( e_g \) volume fractions, and the liquid saturation is defined as \( s_1 = e_1 / e \). Depending on saturation, and also to some degree on wettability, two-phase flow can exist in three possible regimes [28].

In PEMFC electrodes, the capillary number (the ratio of viscous forces to interfacial tension forces) is generally small and liquid transport is dominated by capillary diffusion. Extending classical relationships for the hydrodynamics of a single-phase in porous media to each phase, the conservation equations for the gas and liquid phases take the form [28]:

\[
\begin{align*}
\frac{\partial (1-s) \rho_g}{\partial t} + \nabla \cdot (\rho_g u_g) &= \dot{S}_g, \\
\frac{\partial s \rho_l}{\partial t} + \nabla \cdot (\rho_l u_l) &= \dot{S}_l,
\end{align*}
\]

where \( u_g \) and \( u_l \) are the gas and liquid phase superficial velocities, \( (1-s)e \) and \( s e \) in the left-hand sides represents the volume of each phase, and \( \dot{S}_g \) and \( \dot{S}_l \) are volumetric sources of gas and liquid.

Similarly, extending the single-phase momentum equation to a two-phase system yields

\[
\begin{align*}
u_g &= -\frac{k_g}{\mu_g} \nabla P_g, \\
u_l &= -\frac{k_l}{\mu_l} \nabla P_l,
\end{align*}
\]

where \( \nabla P \) is the pressure gradient, \( (k_g) \) and \( (k_l) \) are phase specific permeabilities, obtained by correcting the bulk permeability \( (k) \) for the effect of the reduced area open to each phase due to the presence of the other phase. With five variables \( (s, u_g, u_l, P_g, P_l) \) and only four equations, an additional relation is required to close the system. This can be obtained by considering the key phenomenon of capillarity associated with interfacial tension. The microscopic capillary pressure is directly proportional to the interfacial tension \( (\sigma) \) and inversely proportional to the radius curvature of the interface \( (r) \), i.e., \( P_c \propto \sigma / r \).
A schematic illustration of capillary motion in a pore is shown in Fig. 7. At the end of the pore where the liquid radius is smaller (lower local saturation), the capillary pressure is greater than at the end with the larger liquid radius (greater local saturation). Because the liquid pressure is the sum of the capillary pressure and the gas pressure \( P_{\text{c}} = \frac{P_{\text{l}}}{C_0} \), the hydrodynamic pressure of the liquid is greater at the end of the pore with the smaller radius. Therefore, the bulk motion of the liquid is toward the end with the greater radius (and local saturation). The transport of liquid water from low to high saturation, as shown in Fig. 7, is counter-intuitive, and illustrates the tendency for water, in hydrophobic media such as those used for gas diffusion layers, to move to ever-increasing pore diameters according to the capillary pressure’s inverse proportionality to the liquid radius. This analysis will be instructive in the subsequent interpretation of visualizations and simulations of flow between the fibres that make up the structure of a GDL.

Introducing the definition of macroscopic capillary pressure into the momentum equations yields:

\[
egin{align*}
    u_{g} &= -\frac{k_{g}}{\mu_{g}} \nabla P_{g}, \\
    u_{l} &= -\frac{k_{l}}{\mu_{l}} \nabla P_{g} - \frac{k_{l}}{\mu_{l}} \nabla P_{c}. 
\end{align*}
\]  

(4)

The last term in Eq. (4b) represents capillary diffusion. Assuming the capillary pressure gradient depends solely on the saturation gradient, the liquid phase momentum equation can be rewritten as

\[
u_{l} = -\frac{k_{l}}{\mu_{l}} \nabla P_{g} - D(s_{l}) \nabla s_{l},
\]  

(5)

with the capillary diffusivity defined as

\[
D(s_{l}) = \frac{k_{l}}{(dP_{c}/ds_{l})}.
\]  

(6)

Eqs. (4) are the basis of the most general multi-fluid formulation used so far in complete fuel cell models [29], and a hierarchy of simplifications, ranging from the mixture model [30] to simple porosity correction schemes have been proposed [28]. One of the central problems in all models of two-phase transport in GDLs is the determination of a constitutive relation for capillary pressure as a function of saturation, the expected general form of which is illustrated in Fig. 8. The prescription of the constitutive relation has so far relied on the so-called Leverett J-function, specifically in the form proposed by Udell [31]:

\[
J(s) = \frac{P_{c}}{\sigma} \left( \frac{k}{\varepsilon} \right)^{1/2},
\]  

(7)

with

\[
J(s) = 1.417(1 - s) - 2.120(1 - s)^{2} + 1.263(1 - s)^{3}. 
\]  

(8)

This is then used to compute the capillary pressure in conjunction with the contact angle \( \theta \)

\[
P_{c} = \frac{\sigma \cos(\theta)}{(k/\varepsilon)^{1/2}} J(s)
\]  

(9)

and in turn to determine the capillary diffusivity.

There are a number of issues with Udell’s relation [28], not the least of which is that the experimental data upon which it is based was obtained in porous samples of soils, which have a radically different structure from fibrous media used in GDLs.

Another key issue in the application of two-phase transport models is the determination of the phase specific permeabilities. These must be corrected for the volume fractions occupied by different phases. One of the
approaches we have recently used to determine the functional dependence of relative permeability and capillary pressure on saturation is based on stochastic pore network model simulations illustrated in Fig. 9. These simulations consist of building a network of pores and throats, with distributions of pore radii corresponding to the porous media of interest, and then solving the momentum transport equations to account for capillary dominated drainage and cluster formation as a function of invading phase saturation. Statistical analysis of the stochastic realizations allows determination of, e.g., the variation of the relative phase permeability and of capillary pressure as a function of saturation.

A more fundamental approach consists of resolving the two phase transport by performing direct numerical simulations that explicitly resolve the flow, including dynamic interface tracking, between the fibres. The equivalent 2D reconstruction of a fibre array corresponding to a typical gas diffusion layer is shown in Fig. 10. The simulation in this systems start with a medium in contact with a liquid water reservoir at the bottom at a higher pressure. A volume of fluid (VOF) technique in conjunction with sub-models for surface tension and wall adhesion is used to track the evolution of the two phase flow, including the interface. The instantaneous pressure field obtained from such simulations, Fig. 11, shows clearly the two-phase interface. 2D, as well as 3D simulations provide fundamental understanding of the two-phase transport, and parametric simulations that directly account for detailed features, such as degree of hydrophobicity, can yield data for determining capillary functions that are more representative of GDLs than Eq. (8).

In recent studies of diffusion layers and two-phase fuel cell models [32,33], it was proposed that liquid water in a GDL evolves in an “inverted tree” structure; i.e. a high number of small capillaries disperse evenly within the GDL and eventually converge into larger capillaries and eventually result in one very large capillary that breaks through the surface. Analysis of the VOF simulations together with recent quantitative visualization experiments performed with laser induced fluorescence [34] suggest fundamentally different transport dynamics.

For a hydrophobically treated GDL, assuming well dispersed PTFE coating (i.e. uniform contact angle), the transport within the GDL is a process of pressure buildup and breakthrough, due to the fact that capillary forces provide the main resistance to the progression of the interface in the fibrous hydrophobic structure. As water is produced in the catalyst layer, the pressure in the liquid increases, and the fluid surface slowly passes through the constrictions of the cross-sections generated by intersecting fibers (see Fig. 12). When the water has passed halfway through each constriction, the curvature of the surface decreases due to the expanding radius of the surface. Thus, the liquid pressure at the interface drops and induces hydrodynamic flow in the direction of the expanding surface. Subsequently, the liquid expands rapidly above this cross-section until the fluid interface contacts the next set of fiber intersections. The process then begins again. The fluid will preferentially pass through the fiber cross-sections featuring the greatest spacing, as this reduces capillary pressure resistance. A schematic of the overall flow mechanism inferred from the analysis of VOF simulations and quantitative visualization [34] is illustrated in Fig. 13, showing a process dominated by fingering and
channeling, and featuring numerous “dead ends” where water transport recedes when an adjacent breakthrough channel forms.

3. Integration into CFD-based fuel cell models

One of the most challenging aspects of computational modelling of PEMFCs is the multi-physics nature of the transport processes, and the coupling between these processes as illustrated in Fig. 14. Each circle in the figure represents a transport equation governing a particular process, and the arrows pointing outward represent the effects of a given transport process on other equations. The transport equations for gas species, water in liquid and solid (membrane) phases, and energy are closely coupled with all other conservation equations. Among all the
processes, the transport of water, which exists in the system in the forms of vapour, liquid, and absorbed in the polymer, is central to all other transport processes. Unsaturated water in the vapour phase affects local relative humidity in the vicinity of the membrane and thus water transport across the membrane and protonic conductivity, which in turn impacts the electric potential field. The transport of liquid water in the porous media affects mass transport of gas species, while liquid water in the gas channels changes the pressure field and may alter the overall flow distribution in a unit cell. A key aspect of any simulations involving two-phase transport and phase change is the proper resolution of heat transfer and irreversibilities [35].

Fig. 15 illustrates the predicted distribution of the rate of phase change and of liquid water saturation in the cathode of a PEMFC using a 3D coupled implementation of the multi-fluid model presented earlier. In this figure, the top and bottom of the vertical axis correspond to the catalyst layer/GDL and gas flow channel/GDL interfaces, respectively, and the planes correspond to successive locations from the inlet to the outlet of the fuel cell section. Positive phase change values correspond to evaporation, which can be found under the land areas due to the increased pressure drop. The pressure drop, an artifact of the increased resistance to gas transport below the land area, reduces the vapour pressure below the saturation point. Negative values, indicating condensation, are most prevalent at the catalyst layer/GDL interface because of the oxygen consumption and the production of water vapour. The corresponding distribution of liquid water saturation in the cathode’s GDL, shown on the right-hand side, indicates maximum saturation levels of about 10% under the land area at the end of the channel.

Transport of water across the membrane links, as noted earlier, the anode and cathode sides of a fuel cell. Polarization curves obtained using a complete fuel cell model that incorporates the binary friction membrane model (BFM2) discussed in Section 2.1 are presented in Fig. 16. A particularly important issue here is the coupling...
of water transport within the membrane to water transport in the electrodes in such a way that mass conservation is ensured. An empirical relationship between water content and water activity (membrane sorption isotherm) has to be enforced to this end. Fig. 16 compares the effects of different humidification regimes on the anode and cathode sides (25% and 75% relative humidity). The Springer model predicts similar polarization curves for both regimes, while the BFM2 predicts a substantially larger loss for the RH = 25/75 regime (dry anode). An analysis of the source of this loss reveals that it is primarily due to larger ohmic losses predicted through the membrane as a result of drying on the anode side. In this case, there is no water to hydrate the anode catalyst layer, unlike the case of the dry cathode (RH = 75/25), where the water production from the oxygen reduction and the cathode-to-anode back-diffusion helps to maintain a more uniform membrane saturation. This highlights the crucial importance of membrane modelling, particularly for fuel cell systems that rely on passive water management, such as air breathing cells considered next.

Exploitation of microscale transport processes in conjunction with macro-manufacturing makes it possible to conceive of very high power density fuel cells [3], which also have the potential of being significantly cheaper than planar plate and frame fuel cells. Micro-fabrication of flow fields, current collectors, and electrical interconnects have been used in a number of designs, but these have generally relied on the traditional planar MEA architecture. Non-planar designs can achieve much higher active area to volume ratios, and hence higher volumetric power densities, as was demonstrated in our waved cell topology [36]. For applications requiring low power, operation in passive air-breathing mode offers several advantages, including system simplification resulting from elimination of manifolds and ancillaries such as fans and increased volumetric power density. Mass transfer limitations are however an issue in such designs [37].

Computational modelling has been particularly valuable in analyzing a concept that exploits the microscale transport principles in conjunction with an air-breathing non-planar patented design illustrated in Fig. 17. The micro-structured electrodes in this design consist of a nano-porous material featuring mean pore diameters on the order of 100 nm, i.e. two orders of magnitude smaller than in typical GDLs (Fig. 6). With comparable levels of porosity, the nano-porous GDL can increase the active area of the deposited catalyst by exploiting the surface to volume ratio [3]. The higher functionality of such a GDL can be impeded by [37]:

- the inability of nano-scale porous media to effectively transport reactant and product gases to the entire active area; and
- the limited ability of natural convection to passively regulate temperature and supply adequate amounts of oxygen to the cathode.

The CFD-based tools were adapted to this problem [37] to assess the functionality of the design, determine the severity of potential transport limitations, and to identify design parameters to alleviate these. In addition to the coupled transport processes discussed earlier, two specific process that have to be accounted for in this case are natural convection in the ambient air, and Knudsen diffusion though the GDL.

Fig. 18 illustrates the distribution of oxygen for a system consisting of three adjacent electrodes subjected to non-uniform convective heat and mass transport currents at the top. The catalyst layer is located along the left hand vertical axis for each electrode. A minimum oxygen mass fraction of 0.089, a 61% drop from the ambient, is obtained in the central cathode. Though the reduction in oxygen concentration is significant, the fuel cell is still far from being oxygen starved, indicating that the GDL...
parameters and dimensions considered will not result in insurmountable mass transport limitations.

4. Concluding perspective

Fuel cells are at the vanguard of emerging clean energy technologies, but must surmount a number of key challenges in order to successfully compete against incumbents such as IC engines and batteries. The availability of reliable and functional computational tools could help achieve dramatic reductions in development cycles and costs, and could spur innovative design required to surmount some of the cost and performance challenges. In this paper we have reviewed the array of transport phenomena encountered in PEM fuel cells, and presented some of the progress made in our laboratory in addressing some of the fundamental issues that still limit the capabilities of computational fuel cell engineering (CFCE).

A new model for coupled protonic and water transport in polymer membranes was presented. This general model is based on rational and physical consideration, removes several of the limitations of the empirical models used to date, and should provide a good foundation for modelling all perfluoro-sulfonic acid membranes. Approaches combining pore network modelling, direct numerical simulation of two phase flow in reconstructed gas diffusion layers, and quantitative visualization have been described, and the sample results presented revealing fundamental mechanisms of liquid water transport in hydrophobic GDLs. The important issue of integration and coupling was also discussed in the context of CFD-based fuel cell models, and we have shown that used judiciously, current models and computational tools can be used in both conventional designs and in novel micro-structured fuel cells to provide detailed information and to analyze design features.

Numerous outstanding problems need to be resolved before truly functional computational fuel cell engineering capabilities are achieved. The broader applicability of the multi-fluid model comes at the cost of solving for an additional set of field equations and the required coupling of the phases. This makes the numerical solution less stable and more computationally intensive. Alternative models based on various levels of simplifications should be better assessed, and their range and regimes of validity fully established in order to allow multiple levels of modelling and practical simulations, particularly for complete cells and stacks. In any case a critical issue for all two-phase models is the determination of appropriate transport parameters for the various electrode materials (including the catalyst layer), and in particular the relative permeability and the capillary pressure function.

One of the inherent advantages of the Binary Friction Membrane Model is that it can be reduced to represent condition under which AC impedance measurements are conducted, thus allowing determination of some of the transport parameters using conductivity data. Experiments and fundamental micro/nanoscale simulations, such as molecular dynamic simulations, are required to determine the fundamental properties of specific PFSA membranes, including the diffusion and interaction coefficients, and the sorption isotherms. Further this model needs to be extended to liquid water equilibrated membranes and to account for transients.

PEMFC catalyst layers consist of a complex composite structure of interconnected, microporous agglomerates surrounded by macropores. Charged species, heat, liquid water, and gas all flow through this layer. Modelling and experimental characterization of this layer is one of the most difficult problems in PEMFCs. Multi-scale modelling strategies are required to resolve the catalyst structure and guide the formulation of improved macroscopic models suitable for use in a CFCE framework.

The functionality and usefulness of computational fuel cell models in a design environment will require the effective and robust integration of the various submodels in a CFCE framework that will allow not only design analysis, but also systematic optimization. This will necessitate the development of novel algorithms that take into account the specific nature of the couplings, the large range of scales encountered in PEMFCs fuel cells, and the multi-variable nature of the optimization problems.

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