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Polymer electrolyte fuel cell transport mechanisms: simulation study of hydrogen crossover and water content

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ABSTRACT

Hydrogen crossover and membrane hydration are significant issues for polymer electrolyte fuel cells (PEFC). Hydrogen crossover amounts to a quantity of unspent fuel, thereby reducing the fuel efficiency of the cell, but more significantly it also gives rise to the formation of hydrogen peroxide in the cathode catalyst layer which acts to irreversibly degenerate the polymer electrolyte. Membrane hydration not only strongly governs the performance of the cell, most noticeable through its effect on the ionic conductivity of the membrane, it also influences the onset and propagation of internal degradation and failure mechanisms that curtail the reliability and safety of PEFCs. This paper focuses on how hydrogen crossover and membrane hydration are affected by; (a) characteristic cell geometries, and (b) operating conditions relevant to automotive fuel cells. The numerical study is based on the application of a general transport equation developed previously to model multi-species transport through discontinuous materials. The results quantify (1) the effectiveness of different practical mechanisms which can be applied to curtail the effects of hydrogen crossover in automotive fuel cells and (2) the implications on water content within the membrane.

INTRODUCTION

Transportation plays a pivotal role in governing the performance of polymer electrolyte fuel cells (PEFC). Cell performance in the form of polarisation curves indicate that cell potential decreases from its thermodynamic equilibrium potential with increasing current density because of irreversible losses. There are generally five sources that contribute to these irreversible losses; (a) the activation polarisation; (b) the mass transportation polarisation; (c) the Ohmic polarisation, and; (d) internal currents which induce efficiency losses. The movement of and cross-interaction between constituent species entering and leaving the cell through the anode and cathode gas diffusion layers (GDL) ultimately controls the magnitude of each of these loss mechanisms. Therefore, in order to reduce these losses and from a technical perspective to bring fuel cells closer towards key performance targets such as power

densities in the 1 kW/kg region and survivability through 5,500 hrs, there is a need to improve our understanding of the underlying phenomenological transport processes.

Over the past two decades water transportation has become a dominant aspect of PEFC research and development [1-4]. Fundamentally, the polymer membrane requires water during operation to allow protons to migrate from the anode to the cathode. Too little water results in membrane dehydration, causing the Ohmic resistance of the membrane to rise. Membrane dehydration can be precipitated particularly by an electro-osmotic drag process where protons travelling through the membrane from the anode catalyst layer to the cathode catalyst layer forcibly drag water molecules towards the cathode (Fig. 1). Since no water is produced at the anode, it can be supplied externally in order to limit dehydration. Contrary to dehydration propagating from the anode side, water which forms in the cathode catalyst layer due to oxygen reduction can accumulate and condense to impede the transport of oxygen to the catalyst sites. In reality it is very likely that dehydration and flooding will co-exist within operational fuel cell stacks and that careful configuration and prudent operation can enable both phenomenological processes to be suitably managed to counterbalance one another [5]. Flooding and dehydration can also precipitate degradation and failure mechanisms within cells through less obvious internal processes. However, these mechanisms can also be controlled if water management is better understood [6].

A vast number of models have been published that describe the physical behaviour of water within a fuel cell [3, 4]. Overall, these models can be classed according to the 'benchmark' approaches that they adopt to describe water transport. These benchmark models have been developed by the following research groups;

- A. Dilute Solution Theory
 - 1. Springer et al [7]
 - 2. Bernardi and Verbrugge [8, 9, 10]
- B. Concentrated Solution Theory
 - 3. Fuller and Newman [11, 12, 13]

In our previous work, we established a validated general transportation equation that describes the physical behaviour of a constituent species within a fluid mixture and which also provides a theoretically consistent meaning for the benchmark fuel cell modelling approaches listed above [14, 15]. The general transport equation was applied to simulate transport across the membrane as an inter-dependant, simultaneous four-component system comprising of water, hydrogen ions, hydrogen and electrolyte [15].

The characterisation of hydrogen crossover and non-uniform water distributions across the cell membrane for fuel cells under load remains to be a critical area of research. Computational methods are continually shedding light on how the fluid dynamics within the gas channels and gas diffusion layers can be optimised for water balance [16, 17, 18]. However, these computational methods cannot be readily applied to model transport across the membrane because fundamentally, the membrane does not have a well-definable regular, rigid, porous structure and because it is an acidic electrolyte with fixed charge groups which interact with positively charged species, i.e., protons. Critically, since the membrane influences cross-transport to/from the anode and cathode channels via the gas diffusion layers, it is necessary to have a firm understanding of how the behaviour of multi-species mixtures change according to thermodynamic conditions in the adjacent cell regions, such as the gas channels, the gas diffusion layers and the catalyst layers. The purpose of the current study is to apply the previously developed general transport equation to further assess how membrane thickness, reactant feed humidification temperature and applied pressure differential all influence both hydrogen permeation and water distribution through the membrane.

Figure 1: Fuel Cell Processes

MODEL APPLICATION

A summary is provided here of the key equations and assumptions applied in the current model [15].

GENERAL TRANSPORT EQUATION

The first component of the general transport equation is the driving force equation, which accounts for the physical conditions that drive intermolecular transport;

$$d_i = \frac{c_i}{c_T} \frac{1}{RT} \left[\nabla \mu_i + \bar{S}_i \nabla T - \frac{M_i}{\rho} \nabla p - \left(X_i - \frac{M_i}{\rho} \sum_{j=1}^v c_j X_j \right) \right] \quad [1]$$

An electric potential field set up by an electric current can cause a species with zero valence to be electro-osmotically dragged, due to the flux of hydrogen ions. Correspondingly, it is possible to define the molar force X_i as follows;

$$X_i = \xi_i \dot{n}_{H^+} \frac{RT}{D_{i,m}} \frac{c_m}{c_T c_i} \quad [2]$$

The second component to the general transport equation is the molecular and thermal diffusion equation;

$$d_i = \sum_j \frac{c_i c_j}{c_T^2 D_{ij}} \left[v_j - v_i + \left(\frac{D_j^T}{\rho_j} - \frac{D_i^T}{\rho_i} \right) \nabla \ln T \right] \quad [3]$$

Substituting equation (1) into (3) for d_i yields the general transport equation;

$$\begin{aligned} c_i \left[\nabla \mu_i + \bar{S}_i \nabla T - \frac{M_i}{\rho} \nabla p - \left(X_i - \frac{M_i}{\rho} \sum_{j \neq i}^v c_j X_j \right) \right] \\ = RT \sum_j \frac{c_i c_j}{c_T D_{ij}} \left[v_j - v_i + \left(\frac{D_j^T}{\rho_j} - \frac{D_i^T}{\rho_i} \right) \nabla \ln T \right] \end{aligned} \quad [4]$$

Overall, the general transport equation considers intermolecular transport due to:

- a gradient in electrochemical potential, composed of;
 - a gradient in species concentration, and
 - a gradient in electric potential
- an overall temperature gradient
- a gradient in total pressure
- a force induced by an external field

MODEL ASSUMPTIONS

General Assumptions

In the current work, the general transport equation is applied to model vapour-phase multi-species transport in 1D across a single cell. The assumptions of the current model are identical to the previous model [15] in all aspects but one; for the current study, it is assumed that the cell can operate under non-isobaric conditions. The auxiliary equations employed in the current study are also identical to those previously employed [15].

Results for the current study are presented in three stages. First, we validate the model by comparing the calculated H_2 crossover rates to measured values from literature [19] at nominal load conditions. Second, we advance the model to analyse the suppression of hydrogen crossover in the context of three key parameters: (1) membrane thickness, (2) applied pressure differential and (3) operating current density. Finally, we consider the impact of these same parameters and of the fourth key parameter, the reactant humidification temperature, on net water transport and water distribution within the membrane.

Validation Assumptions

In order to validate the model, we employ the data recently published by Cheng et al [19]. They experimentally characterised the hydrogen crossover rate within a 4.4cm^2 experimental fuel cell as a function of anode backpressure. The hydrogen crossover rate was then applied to the following equation to determine the hydrogen permeability coefficient (mol/cm-atm-s):

$$\psi_{H_2}^{perm} = \frac{I_{H_2}^{cross} t_{mem}}{p_{H_2}^{CH-A}} \quad [5]$$

In the current model, we calculate the hydrogen permeability coefficient as a function of the net flux ratio of hydrogen α_{H_2} which is determined through a convergence process as detailed in our previous work [14, 15]. The hydrogen permeability coefficient is calculated as;

$$\psi_{H_2}^{perm} = \frac{J \alpha_{H_2} t_{mem}}{2F p_{H_2}^{CH-A}} \quad [6]$$

where J is the current density (A/cm^2), t_{mem} is the membrane thickness (cm), $p_{H_2}^{CH-A}$ is the partial pressure of hydrogen in the anode channel (atm) and F is the Faraday constant (C). The hydrogen partial pressure in the anode channel is calculated as [19];

$$p_{H_2}^{CH-A} = \left(\frac{p_{H_2}^{IN-A}}{p_{total}^{IN-A}} \right) \left(\frac{p_{total}^{IN-A} + p_{backpressure}}{2} \right) \quad [7]$$

In the current study, we validate our calculated permeability coefficients against those results presented by Cheng et al [19] for a cell employing a $50\text{ }\mu\text{m}$ thick polymer electrolyte membrane operated with fully humidified reactant supply streams at 80°C , 100°C and 120°C , and with anode backpressures at 2.02 atm and 3.04 atm.

SIMULATED CONDITIONS

In the context of operational fuel cells, the four key mentioned parameters can be easily manipulated and in the context of the current study enable a straightforward assessment of how hydrogen crossover can be suppressed and membrane water content can be improved.

Membrane Thickness

In the literature, it has been established that thin ($\approx 50\text{ }\mu\text{m}$) membranes remain well hydrated at all current densities and correspondingly exhibit high proton conductivity compared to thick membranes ($\approx 175\text{ }\mu\text{m}$) [7]. This owes to the fact that thinner membranes imply a shorter transport path for water from cathode to anode, inherently mitigating electro-osmotic drag. However, thin membranes are more susceptible to failure due to the formation and propagation of pinholes [6]. For longevity, therefore, especially for applications where the load profile involves repeated stop-start cycles, it can be more favourable to opt for thicker membranes. Arguably, longer transport paths can also suppress hydrogen crossover. Two membrane thicknesses are considered in the current study; $50\text{ }\mu\text{m}$ and $175\text{ }\mu\text{m}$.

Applied Pressure Differential

Non-isobaric conditions can induce flow in the form of forced convection from regions of high pressure to regions of low pressure. In addition, since the concentration of multi-component gas constituents increases with gas pressure, diffusive transport from regions of high concentration to regions of low concentration can also be enhanced. Pressure differentials could therefore be applied to enable the internal humidification of cells using water that is generated in the cathode catalyst layer. It is noteworthy, however, that prolonged exposure to pressure differentials in the region of 1 – 2 atm can hasten the onset of membrane failure. Three pressure differentials are currently considered;

- $dp = 1.00\text{ atm}$ ($p_A = 2.50\text{ atm}$, $p_C = 3.50\text{ atm}$)
- $dp = 0.50\text{ atm}$ ($p_A = 2.75\text{ atm}$, $p_C = 3.25\text{ atm}$)
- $dp = 0.00\text{ atm}$ ($p_A = 3.00\text{ atm}$, $p_C = 3.00\text{ atm}$)

Operating Current Density

As the flux of protons through the membrane increases in direct proportion to the current density, the electro-osmotic drag of water from anode to cathode also

increases correspondingly. As such, electro-osmotic drag can become increasingly prominent at high current densities and can counteract any water transport from the cathode to the anode. The current study will consider how water content within the membrane is affected by applied pressure differentials under a range of operating current densities (0.4 – 1.6 A/cm²).

Reactant Feed Humidification Temperature

The amount of water supplied to a cell through the reactant feeds can be controlled as a function of humidification temperature; since saturation vapour pressure increases exponentially with gas temperature, the amount of water vapour that is supplied to a cell therefore also increases when reactant gases are fully humidified [7]. The reactant humidification temperature therefore provides a means by which hydration at the membrane boundaries can be controlled and consequently the water content within the membrane itself. In addition, other gas and vapour properties that govern transport such as diffusivity and permeability also have strong dependencies on temperature. The current study considers the effect of fully humidifying the reactant feeds (100% relative humidity) to a range of humidification temperatures (80 – 120°C).

RESULTS AND DISCUSSION

HYDROGEN CROSSOVER – MODEL VALIDATION

The data in Table 1 compares the calculated hydrogen permeability coefficients obtained from the general transport equation to the experimentally-derived data published by Cheng et al [19].

Humidification Temperature (°C)		Hydrogen Permeability Coefficient (mol/cm-atm-s)	
		Anode backpressure	
		3.04 atm	2.02 atm
80	Cheng et al [19]	3.87E-11	3.71E-11
	General transport equation	1.91E-12	1.15E-11
100	Cheng et al [19]	6.13E-11	5.24E-11
	General transport equation	1.90E-11	6.38E-11
120	Cheng et al [19]	1.04E-10	1.08E-10
	General transport equation	1.36E-10	4.23E-10

Table 1: Calculated and experimentally-derived hydrogen permeability coefficients for 50 µm polymer electrolyte membrane

In general, Table 1 demonstrates that the hydrogen permeability coefficient increases with increasing humidification temperature. The same observation can be deduced from the work reported previously by Weber et al [20]; their data fit suggests that for a polymer membrane with a water volume fraction of 50%, raising

the cell temperature from 80°C to 120°C will have the effect of increasing the hydrogen permeability coefficient from 4.52 to 9.36 mol/cm-atm-s. The phenomenon can be attributed to the increase in hydrogen diffusivity at higher operating temperatures.

Overall, the calculated hydrogen permeability coefficients are generally within the same order of magnitude as the experimentally-derived values. The two sets of results do not clearly elucidate, however, the dependence of hydrogen crossover on both anode and cathode channel pressures. In order to do this, the current general transport equation is applied to simulate hydrogen crossover when the pressure differential across the cell is 0.5 atm and 1 atm respectively.

HYDROGEN CROSSOVER – SUPPRESSION MECHANISMS

Figures 2 and 3 show the effect of pressure differentials across the cell on the net flux ratio of hydrogen as a function of operating current density. Figures 2 and 3 both demonstrate that the net flux ratio generally decreases with current density. It is also evident that raising the pressure differential across the cell does indeed suppress hydrogen crossover at all simulated operating current densities. In Figure 2, the net flux ratio decreases by 12% from 0.0326 to 0.0287 at 0.4 A/cm² and by 11% from 0.0107 to 0.0095 at 1.2 A/cm² when the pressure differential across the cell is increased from 0.5 atm to 1 atm. Figure 3 shows the change in the net hydrogen flux ratio for the 175 µm membrane. In the first instance, it is possible to see that thicker membranes inherently suppress the net hydrogen flux ratio by one order of magnitude. At 1 A/cm², a 50 µm membrane achieves a net flux ratio of 0.0114 with a 1 atm pressure differential, whereas the 175 µm suppresses this by 68% to 0.0037. The general reduction in the net hydrogen flux ratio by raising the pressure differential from 0.5 atm to 1 atm for the 175 µm membrane is consistently around 10% for all the simulated current densities.

Figure 2: Net hydrogen flux ratio as a function of operating current density; membrane thickness: 50 µm, reactant humidification temperature: 80 °C

Figure 3: Net hydrogen flux ratio as a function of operating current density; membrane thickness: 175 μm , reactant humidification temperature: 80 $^{\circ}\text{C}$

MEMBRANE WATER CONTENT

Isobaric Operating Conditions

Water content within the membrane is commonly defined as the number of water molecules per membrane charge site, denoted λ . In the case of Nafion, for example, a charge site is an SO_3^- end group. In previous studies it was shown that when a cell is operated under isobaric conditions and when both reactant feeds are fully humidified, the membrane water content is at a maximum adjacent to the cathode catalyst layer and diminishes towards the anode catalyst layer [7, 15]. This can be observed in Figs. 4 and 5 for a 50 μm membrane and 175 μm membrane respectively when the humidification temperature is 80 $^{\circ}\text{C}$. In both cases, for the current densities simulated the results show that the membrane water content is typically between 14 and 16 per charge site at the cathode interface (thickness fraction = 1). This is consistent with the calculated results for the vapour-phase model presented by Springer et al [7]. The 50 μm membrane maintains a higher water content at all current densities in comparison to the 175 μm membrane. At 1.6 A/cm², the water content at the anode interface is simulated to be 6 molecules per charge site for the 50 μm membrane, whereas for the 175 μm membrane it is much lower at around 1 molecule per charge site.

Figures 6 and 7 show the simulated influence of humidification temperature on water content as a function of membrane thickness fraction for the two thicknesses considered. The humidification temperature is now raised to 110 $^{\circ}\text{C}$, which again increases the water content of the reactant feeds. The noticeable difference is that the uniformity in water content is slightly improved throughout the thickness of the membranes for both cases. For the 50 μm membrane, the water content per charge site at the anode interface at 1.6 A/cm² is now 7 molecules per charge site and now approaching 3 molecules per charge site for the 175 μm membrane.

Figure 4: Membrane water content as a function of thickness fraction ($dp = 0 \text{ atm}$); membrane thickness: 50 μm , reactant humidification temperature: 80 $^{\circ}\text{C}$

Figure 5: Water content as a function of membrane thickness fraction ($dp = 0 \text{ atm}$); membrane thickness: 175 μm , reactant humidification temperature: 80 $^{\circ}\text{C}$

In general, Figs. 4 – 7 also show that the difference in water content between the anode and cathode interfaces increases with current density. Raising the current density has the effect of increasing the electro-osmotic drag, thereby reducing water content throughout the membrane and in particular drying out the side of the membrane closest to the anode catalyst layer.

Figure 6: Water content as a function of membrane thickness fraction ($dp = 0 \text{ atm}$); membrane thickness: 50 μm , reactant humidification temperature: 110 $^{\circ}\text{C}$

Figure 7: Water content as a function of membrane thickness fraction ($dp = 0$ atm); membrane thickness: $175 \mu\text{m}$, reactant humidification temperature: 110°C

Non-Isobaric Operating Conditions

From here, it is possible to consider the effect that an applied pressure gradient across a cell has on water distribution. The increase in water distribution is considered, $\Delta\lambda$, which is the difference between the water content at the cathode-membrane and anode-membrane interfaces (increase in water content is implied in the direction of cathode from the anode).

Figures 8 and 9 show the increase in water content for a $50 \mu\text{m}$ membrane supplied with fully humidified reactant gases at 80°C and 110°C respectively. In general, the results show that $\Delta\lambda$ increases with current density. This general phenomenon can also be observed in Figs. 12 and 14 for a $175 \mu\text{m}$ membrane. What is most significant about Figs. 8 and 9 however is that it shows that the difference in water content can be suppressed when both the pressure differential across the cell and the reactant humidification temperature are increased. Figure 8 demonstrates that for cells operating with reactant feeds fully humidified at 80°C , the pressure differential alone is not necessarily sufficient to enable a more uniform water distribution profile across the membrane from cathode to anode. Figure 9 for 110°C , however, infers that the higher water content at the membrane boundaries causes the increase in water content across the membrane to be suppressed under both isobaric and non-isobaric conditions. When $dp = 1$ atm the increase in water content is suppressed further.

Figures 10 and 11 can be compared to Figs. 4 and 5 respectively to understand why a dp of 1 atm causes a higher $\Delta\lambda$ at 80°C and a lower $\Delta\lambda$ at 110°C for the $50 \mu\text{m}$ membrane compared to when dp is equal to 0. At 80°C , Fig. 10 shows that the applied pressure differential makes only a marginal difference in water content and only noticeably increases the water content at the cathode interface; at 1.6 A/cm^2 , the water content rises from 15.7 to 16.1 whereas for the anode interface, the water content remains at around 5.9 . It suggests that an applied pressure differential at low humidification temperatures only serves to marginally increase the overall water content and at best only noticeably affects the cathode side. As such, $\Delta\lambda$ is marginally higher when

$dp = 1$ atm compared to when $dp = 0$ atm at 80°C . Figure 11 in comparison to Fig. 6 shows the opposite effect; the increase in humidification temperature to 110°C coupled with an applied pressure differential causes the water content at the anode interface to improve whilst closer to the cathode the water content is kept fairly constant. At 1.6 A/cm^2 , the water content at the cathode interface remains at around 14.5 whereas at the anode interface it increases from 7.3 to 7.9 .

Figure 8: Increase in water content across $50 \mu\text{m}$ membrane operated; reactant humidification temperature: 80°C

Figure 9: Increase in water content across $50 \mu\text{m}$ membrane; reactant humidification temperature: 110°C

Figure 10: Water content as a function of thickness fraction ($dp = 1$ atm); membrane thickness: $50 \mu\text{m}$, reactant humidification temperature: 80°C

Figure 11: Water content as a function of thickness fraction ($dp = 1$ atm); membrane thickness: $50 \mu\text{m}$, reactant humidification temperature: 110°C

Figure 12: Increase in water content across $175 \mu\text{m}$ membrane; reactant humidification temperature: 80°C

Figures 12 and 13 demonstrate the effect of using a thicker membrane. The general behaviour observed for the $50 \mu\text{m}$ membrane is seen again for the $175 \mu\text{m}$ membrane. Noticeably, the increase in water content between the anode and cathode ends of the membrane are more significant; at 80°C for example, the maximum increase for the $50 \mu\text{m}$ membrane is around 10 water molecules per charge site, however for $175 \mu\text{m}$ at the same humidification temperature, the increase is 15 water molecules per charge site. This can be attributed to the longer diffusion path for water from the cathode to the anode. Similarly to Fig. 9, Fig. 13 shows that while a higher reactant humidification temperature alone can enhance uniformity in membrane water content, when this is coupled with an applied pressure differential the uniformity in water content can be enhanced further.

Figure 13: Increase in water content across $175 \mu\text{m}$ membrane; reactant humidification temperature: 110°C

In Figs. 12 and 13, higher values for $\Delta\lambda$ are obtained at 80°C and lower values at 110°C when $dp = 1$ atm compared to when $dp = 0$. This is similar to the phenomenon observed for the $50 \mu\text{m}$ membrane. This again can be attributed to the fact that the pressure differential at the lower humidification temperature only serves to marginally increase the water content at the cathode, whereas raising the humidification temperature as well as the pressure differential both ensure the hydration of the anode improves while the cathode water content remains generally the same. This is evident in Figs. 14 and 15 when compared to Figs. 5 and 7. Overall, this suggests that the application of a pressure differential only benefits the membrane in terms of water content if the vapour pressures of the reactant supplies are raised. Humidification to 80°C does not provide sufficient water for the pressure differential to enable any significant effect, whereas humidification to 110°C evidently does.

Figure 14: Water content as a function of thickness fraction ($dp = 1$ atm); membrane thickness: $175 \mu\text{m}$, reactant humidification temperature: 80°C

Figure 15: Water content as a function of thickness fraction ($p = 1$ atm); membrane thickness: 175 μm , reactant humidification temperature: 110 $^{\circ}\text{C}$

CONCLUSIONS

The effect of applied pressure differentials (0 – 1 atm) across a polymer electrolyte membrane fuel cell were simulated in the current study over a range of a range of reactant humidification temperatures (80 to 120 $^{\circ}\text{C}$), two membrane thicknesses (50 and 175 μm) and a range of operating current densities (0.4 – 1.6 A/cm^2).

Predicted values of hydrogen permeability for a cell employing a 50 μm thick membrane operated with reactant feeds fully humidified at 80 $^{\circ}\text{C}$, 100 $^{\circ}\text{C}$ and 120 $^{\circ}\text{C}$ and 3.04 atm and 2.02 atm anode backpressures were compared against experimentally-derived values from literature [19]. The results were generally within the same order of magnitude as the experimentally-derived values and showed that hydrogen permeability increased with cell temperature.

The effect of raising the pressure differential across a working cell was simulated for two membrane thicknesses; 50 μm and 175 μm . The simulated results showed that for the cell operating with the 50 μm membrane and humidified to 80 $^{\circ}\text{C}$, the net hydrogen flux ratio could be suppressed by at least 11% if the pressure differential was raised from 0.5 atm to 1 atm. Similarly, for the cell operating with the 175 μm membrane, the net hydrogen flux ratio could be suppressed by 10% if the pressure differential was increased in the same way.

Simulated results to show the effect of a 1 atm pressure differential on membrane water content revealed that for the 50 μm and the 175 μm membranes, the water distribution could be made more uniform if both the reactant feeds were fully humidified to higher temperatures (i.e., 110 $^{\circ}\text{C}$ in lieu of 80 $^{\circ}\text{C}$). At the lower temperature, the applied pressure differential only served to marginally increase the water content at the cathode interface, whereas at the higher temperature, the pressure differential enabled the water content at the anode interface to increase. This suggests that pressure differentials only cause an effect if the water content of the reactant supplies are raised.

While the work presented here focuses on the effect of cathode pressurisation on hydrogen crossover and membrane water content, it is noteworthy that oxygen crossover and anode pressurisation can also occur. Oxygen crossover can additionally contribute towards a loss in fuel efficiency [20], while anode pressurisation could result in a strong back-diffusion of water. Such phenomenon can be simulated explicitly through the current modelling framework.

Current work is focusing on the amalgamation of the general transport equation with a statistical-based multi-dimensional, multi-phase transport model for the channel sections, gas diffusion layers, catalyst layers and microporous layers of a representative elementary volume (REV). Future work will also focus on validating simulated results against permeability data over a wider range of cell configurations and operating conditions.

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DEFINITIONS, ACRONYMS, ABBREVIATIONS

c_i	concentration of species i
d_i	molecular driving force of species i
D_{ij}	diffusion coefficient of species pair i,j
D_i^T	thermal diffusion coefficient of species i
F	Faraday constant
I_i	molar crossover rate of species i
J	current density
M_i	molar mass of species i
$\dot{n}_i$	molar flux of species i
p	pressure
R	Universal gas constant
$\bar{S}_i$	molar entropy of species i
t_{mem}	membrane thickness
T	temperature
v_i	velocity of species i
X_i	general molar force for species i
Greek	
α_i	net flux ratio of species i
λ	moles of water per mole of charge site
μ_i	molar electrochemical potential of species i
ξ_i	electro-osmotic drag ratio of species i
ρ_i	density of species i
ψ_i^{perm}	permeability coefficient of species i