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1 Anode partial flooding modelling of Proton Exchange Membrane Fuel

2 Cells: Model development and validation

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13 Highlights

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- 15 • A fully coupled 2D, along-the-channel, two-phase flow, non-isothermal, CFD model is
16 developed.
- 17 • Temperature rise due to electrochemical reactions, ohmic resistance and water phase-transfer is
18 analysed.
- 19 • Mathematical expressions of liquid water saturation against current density at anode and cathode
20 are regressed.
- 21 • Relationship between the liquid water saturation at anode and cathode is built.

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33 **Abstract**

34

35 A two-dimensional along-the-channel CFD (computational fluid dynamic) model, coupled with a
36 two-phase flow model of liquid water and gas transport for a PEM (proton exchange membrane) fuel
37 cell is described. The model considers non-isothermal operation and thus the non-uniform temperature
38 distribution in the cell structure. Water phase-transfer between the vapour, liquid water and dissolved
39 phase is modelled with the combinational transport mechanism through the membrane. Liquid water
40 saturation is simulated inside the electrodes and channels at both the anode and cathode sides. Three
41 types of models are compared for the HOR (hydrogen oxidation reaction) and ORR (oxygen reduction
42 reaction) in catalyst layers, including Butler-Volmer (B-V), liquid water saturation corrected B-V and
43 agglomerate mechanisms. Temperature changes in membrane electrode assembly (MEA) and
44 channels due to electrochemical reaction, ohmic resistance and water phase-transfer are analysed as a
45 function of current density. Nonlinear relations of liquid water saturations with respect to current
46 densities at both the anode and cathode are regressed. At low and high current densities, liquid water
47 saturation at the anode linearly increases as a consequence of the linear increase of liquid water
48 saturation at the cathode. In contrast, exponential relation is found to be more accurate at medium
49 current densities.

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57 **Key works:** PEMFC; liquid water; anode flooding; agglomerate model; non-isothermal; CFD

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59 **1. Introduction**

60 Benefiting from high energy efficiency and low emission, PEMFCs (proton exchange membrane
61 fuel cells), play an important role in transiting a carbon intensive economy to sustainable low carbon
62 future [1-4]. In addition, the features of PEMFCs endow a flexibility and scalability for use with
63 batteries and CHP (combined heat and power) system for hybrid automotive and residential use [5-7],
64 as well as energy conversion from winds to electricity [8]. Among the PEMFC family, the medium
65 and low temperature PEMFCs are considered as promising candidates as portable and automotive
66 power sources. However, they can experience certain operating difficulties associated with water
67 transport and flooding during their operation, especially at a high current density [9-11].

68 Three phases of water co-exist in PEMFCs: as vapour and liquid in porous electrodes and channels,
69 and as a dissolved phase absorbed by membrane and ionomer (membrane and ionomer water uptake).
70 Among three phases, dissolved water can migrate between both electrodes through the membrane
71 under the driving forces of EOD (electro-osmotic drag), back diffusion and hydraulic permeation
72 [12-14]. Liquid water can be generated in terms of water vapour condensation and membrane/ionomer
73 desorption when over-saturated. Simultaneously, liquid water is removed from the MEA (membrane
74 electrode assembly) generally by reactant gases flowing along the channels. If water generation rate is
75 faster than the removal rate, excess water will accumulate in the electrodes and flow channels, leading
76 to a water flooding inside the cell. This flooding can reduce the effective porosity in the porous media
77 and increase the pressure drop along the channel, inhibiting the gas transport to the active sites in CLs
78 (catalyst layers), finally resulting in a decline in the cell performance [9, 15, 16]. Water flooding is
79 typically observed at the cathode side due to the fact that water is produced inside the CCLs (cathode
80 catalyst layers) by the ORR (oxygen reduction reaction).

81 There have been numerous studies of water flooding at the cathode but less consideration of water
82 transport related to the anode. However, hydrogen consumption at the anode could also results in the
83 humidified anode reactant gas becoming supersaturated with water vapour, which can result in
84 transfer to liquid water after condensation. Liquid water movement in hydrophilic and hydrophobic
85 anode gas channel, as well as the effect of hydrogen inlet velocity, operating temperature and channel
86 walls wettability, was numerically studied by Ferreira et al. [17]. Experimental studies have detected
87 liquid water in the anode and that more liquid water was observed at a high anode relative humidity
88 [18, 19]. An in-situ detection scheme of anode flooding, developed by O'Rourke et al. [20], showed
89 that anode flooding could be detected prior to a rapid cell voltage decline, which was considered as an
90 early warning of cathode catalyst damage. As reported by Anderson et al. [21], AWR (anode water
91 removal) could be used as a diagnostic tool to assess cathode water flooding in PEM fuel cells.

92 Catalyst layers are complex structures and are a difficult to describe and be fully understood in
93 PEMFCs; not only due to their highly compact structure and complex composition but also because of
94 the coupled electrochemical reactions and transport processes occurred. CLs in PEMFCs facilitate
95 electrochemical reactions and produce water at the cathode. To describe the current density produced
96 on each electrode in fuel cell operation, considering the water generated which can partially cover the
97 platinum catalyst surface, (1-s) corrected B-V equation (s is the liquid water saturation, defining as the
98 volume fraction of void space occupied by liquid water) is usually required instead of the traditional
99 B-V equation [22-24]. The electrochemical activity may be further represented using an agglomerate
100 mode, taking into account the increase in ionomer film thickness surrounding the agglomerate due to
101 ionomer swelling. Furthermore, by considering gas transport resistance and the reduction of porosity

102 due to liquid water occupation, the agglomerate model can also provide a superior representation of
103 the porous CLs in comparison with other models [25, 26].

104 In fuel cell operation, temperature plays a significant role in achieving a high power performance. A
105 variety of parameters, e.g. electrochemical reaction kinetics, mole fractions in gas mixture, electrolyte
106 conductivity, rate of mass transfer, as well as liquid water saturation, are closely correlated with
107 temperature during fuel cell operation. Within PEMFCs, the temperature distribution across the MEAs
108 and along the flow channels can be either estimated by mathematical modelling technique or detected
109 by experimental measurements [27-30]. Ju et al. [27] developed a three dimensional (3D),
110 single-phase, non-isothermal model through a parametric study for GDL thermal conductivity, gas
111 relative humidity and operating cell voltage. Wang et al. [28] measured the temperature profile within
112 a single PEMFC with the aid of ITI (infrared thermal imaging) and found that the downstream
113 temperature is higher than the upstream, and there was an obvious high temperature section in the
114 outlet of oxygen. Temperature measurement were conducted by Zhang et al. [29] and Lin et al. [30]
115 from which they found a correlation between local temperature rise and local current density, and a
116 temperature rise at backsides of both the anode and cathode flow field plates.

117 However, up to now, it is still a challenge to fully couple most aspects mentioned above in
118 modelling endeavor for a highly accurate evaluation of water flooding in a single PEMFC or stack. In
119 this paper, we fully couple seven sub-models and apply them on a two dimensional (2D), ATC
120 (along-the-channel) geometry for a single PEMFC: (1) agglomerate models for gas transport
121 resistance in CLs, (2) two-phase flow models for liquid water saturation, (3) combinational diffusion
122 models for dissolved water transport through the membrane, (4) non-isothermal models for heat
123 transport in MEA and channels, (5) CFD (computational fluid dynamic) models for gas transport

124 along the channels, (6) multicomponent diffusion of reactant gases in gas mixture, and (7) ionomer
125 swelling due to non-uniform distribution of water content. The effect of liquid water on the current
126 density are compared based on three mechanisms, including traditional B-V, (1-s) corrected B-V and
127 agglomerate kinetics. Distributions of reactant gas, liquid water and heat within the cell are
128 investigated. The expressions for the relationship between the liquid water saturation on each
129 electrode and current density, as well as the liquid water saturation between anode and cathode, are
130 regressed.

131 **2. Model description**

132 **2.1 Assumption**

133 Some additional assumptions were employed in this work in addition to those in the previous
134 papers [34-38]:

- 135 1. Steady state, two-phase flow, along the channel model of a PEMFC.
- 136 2. Gas flow in the channels is laminar flow and incompressible.
- 137 3. Reactant gases enter the gas channels in a direction normal to the channel cross section.
- 138 4. Liquid water saturation in channels is continuous.
- 139 5. Gas diffusion layers and catalyst layers are isotropic.
- 140 6. No liquid water enters the channels at the inlets.
- 141 7. Same electrodes and bipolar plates are used in the anode and cathode, respectively.

142 **2.2 Governing equations**

143 The conservation equations of mass, momentum, species, energy and charges are summarized as Eqns.
144 (1) to (4):

$$145 \quad \nabla \cdot (\rho^g \mathbf{u}^g) = S_m \quad (1)$$

$$146 \quad \rho^g (\mathbf{u}^g \cdot \nabla \mathbf{u}^g) = \nabla \left[-pI + \mu^g \nabla \mathbf{u}^g + \mu^g (\nabla \mathbf{u}^g)^T - \frac{2}{3} \mu^g (\nabla \cdot \mathbf{u}^g) I \right] \quad (2)$$

$$147 \quad \rho^g \mathbf{u}^g \cdot \nabla w_i^g - \nabla \cdot [\rho^g \sum_{j=1}^N (1-s') D_{ij} (\nabla x_j^g - w_j^g) \frac{\nabla p}{p} + D_i^T \frac{\nabla T}{T}] = M_i S_i^g \quad (3)$$

$$148 \quad \nabla \cdot [\sum_{i=g,l} (\varepsilon \rho c_p \mathbf{u})_i T] - \nabla \cdot (\sum_{i=g,l,s} k_i \nabla T) = S_T \quad (4)$$

$$149 \quad \sigma_s^{eff} \nabla \cdot \nabla \varphi_s + \sigma_M^{eff} \nabla \cdot \nabla \varphi_M = 0 \quad (5)$$

150 where ρ (kg m⁻³) is density, \mathbf{u} (m s⁻¹) is velocity, p (Pa) is pressure, μ (Pa s) is viscosity, M
151 (kg mol⁻¹) is molecular weight, D (m² s⁻¹) is diffusion coefficient, D^T (kg m⁻¹ s⁻¹) is thermal
152 diffusion coefficient, c_p (J mol⁻¹ K⁻¹) is specific heat capacity, k (W m⁻¹ K⁻¹) is thermal
153 conductivity, T (K) is temperature, σ (S m⁻¹) is conductivity φ (V) is potential. w is mass
154 fraction, x is mole fraction, ε is volume fraction, s' is corrected liquid water saturation and I
155 is identity matrix. Subscript i and j represent species i and j and superscript g means gas phase. S is
156 the source term, which is given in detail in Tables 1 to 3. Note that Eq. (1) to Eq. (3) were applied on
157 reactant gas only and the gas velocity in porous electrodes could be related to pressure according to
158 Darcy's law as below:

$$159 \quad \mathbf{u}^g = -\frac{k_p}{\mu^g} \nabla p \quad (6)$$

160 where k_p (m²) is the permeability of the porous media.

161 For the purpose of describing dissolved and liquid water transport, the following equations were used.

$$162 \quad \nabla \cdot (n_d \frac{i_M}{F}) - \nabla \cdot (D_{w-M} \nabla c_w^d) - \nabla \cdot (\frac{k_{p,M} c_w^d}{\mu_w} \nabla p) = S_w^d \quad (7)$$

$$163 \quad \nabla \cdot (\rho_w^l D_c \nabla s - \frac{\rho_w^l k_r^l \mu_w^g}{k_r^g \mu_w^l} \mathbf{u}^g) = M_w S_w^l \quad (8)$$

164 Eq. (7) was developed by applying the diffusive approach to a conservation equation of dissolved
165 water, while Eq. (8) was obtained by applying the volume average approach to the continuity equation

166 and using Darcy's law for both the liquid and gas phases.

167 The capillary diffusion coefficient, D_c ($\text{m}^2 \text{s}^{-1}$), was calculated using Eq. (9) [31]:

$$168 \quad D_c = -\frac{k_r^l}{\mu_w^l} \sigma \cos(\theta_c) (\varepsilon k_p)^{1/2} \frac{dJ(s)}{ds} \quad (9)$$

169 where k_r^l is the relative permeability of liquid phase, μ_w^l (Pa s) is the dynamic viscosity of liquid
170 water, σ (N m^{-1}) is the surface tension, θ_c ($^\circ$) is the contact angel, ε is the porosity of the electrode,

171 k_p (m^2) is the permeability of the porous electrode, $J(s)$ is the Leverett function.

172 The equilibrium membrane/ionomer water content, determined based on water uptake measurement,
173 is given as [32]:

$$174 \quad \lambda^{eq} = 16.8s + 14.0(1-s) \quad s > 0 \quad (10)$$

175 The permeability of gas channels was calculated by Hagen-Poiseuille equation [34].

$$176 \quad k_{p,ch} = \frac{c_{sf} d_h^2}{32} \quad (11)$$

177 where c_{sf} is the flow shape factor (1.127 for a square cross-section) and d_h is the hydraulic
178 diameter of a channel, which can be calculated by the relation of the cross-sectional area and wetted
179 perimeter.

180 The electrochemical reactions inside the catalyst layers according to three different mechanisms,
181 including B-V, (1-s) corrected B-V and agglomerate assumption, can be written as below:

$$182 \quad i_{BV} = a_{agg}^M i_{0,i}^{ref} \left(\frac{p_i}{c_i^{ref} H_i} \right)^\gamma \left[\exp\left(\frac{-\alpha_{Rd,i} F \eta_i}{RT} \right) - \exp\left(\frac{\alpha_{Ox,i} F \eta_i}{RT} \right) \right] \quad (12)$$

$$183 \quad i_{(1-s)BV} = (1-s) a_{agg}^M i_{0,i}^{ref} \left(\frac{p_i}{c_i^{ref} H_i} \right)^\gamma \left[\exp\left(\frac{-\alpha_{Rd,i} F \eta_i}{RT} \right) - \exp\left(\frac{\alpha_{Ox,i} F \eta_i}{RT} \right) \right] \quad (13)$$

$$184 \quad i_{agg} = nF \left(\frac{p_i}{c_i^{ref} H_i} \right)^\gamma \left[\frac{1}{E_{agg} k_{agg}} + \frac{(r_{agg} + \delta_M + \delta_w)}{r_{agg}} \left(\frac{\delta_M}{a_{agg}^M D_{O_2-M}} + \frac{\delta_w}{a_{agg}^w D_{O_2-w}} \right) \right]^{-1} \quad (14)$$

185 where

$$k_{agg} = \frac{\alpha_{agg}^M i_{0,i}^{ref}}{nF} \left[\exp\left(\frac{-\alpha_{Rd,i} F \eta_i}{RT}\right) - \exp\left(\frac{\alpha_{Ox,i} F \eta_i}{RT}\right) \right] \quad (15)$$

The subscript i is for anode and cathode, respectively, and the superscript γ equals to 0.5 for HOR (hydrogen oxidation reaction) in anode and 1.0 for ORR in cathode. The properties of the catalyst layer (porosity, specific area, ionomer and water film thickness), gas transport through the porous electrode (species diffusion in gas mixture and Knudsen diffusion), numerous key parameters and fundamental equations, were listed in Table 4. Details can be found in literature [34-38].

2.3 Boundary conditions

At anode inlet (A'-B') and cathode inlet (G-H) as shown in Fig. 1, the temperature, mole fractions of reactant gases in gas mixture, and liquid water saturation were given as below:

$$T = T^{cell}, \quad x_{w,a}^0 = \frac{p_{sat} RH_a}{p_a}, \quad x_{H_2}^0 = 1 - x_{H_2O,a,in}, \quad s_a = 0 \quad (16)$$

$$T = T^{cell}, \quad x_{w,c}^0 = \frac{p_{sat} RH_c}{p_c}, \quad x_{O_2}^0 = 0.21(1 - x_{H_2O,c,in}), \quad x_{N_2}^0 = 0.79(1 - x_{H_2O,c,in}), \quad s_c = 0 \quad (17)$$

The gas velocities at the inlets of both electrodes were related to their stoichiometry shown as follow:

$$\mathbf{u}_a^{g,0} = \frac{\xi_a RT i^{ref} A_M}{2F p_a x_{H_2} A_{ch}}, \quad \mathbf{u}_c^{g,0} = \frac{\xi_c RT i^{ref} A_M}{4F p_c x_{O_2} A_{ch}} \quad (18)$$

where i^{ref} is the reference current density (defined as 1.0 A cm⁻² in this study), ξ_a and ξ_c are the stoichiometry of the anode and cathode, respectively. A_M (m²) and A_{ch} (m²) are the effective area of electrode and the cross-sectional area of channel, respectively.

At the inlet and outlet of both the anode and cathode, the pressure was given as a boundary condition.

$$p_a = p_a^0, \quad p_c = p_c^0 \quad (19)$$

The water content on the CL-membrane interfaces of anode (C-C') and cathode (F-F') were defined as Dirichlet boundaries with the values according to the following equations [14, 36-38]:

206 $\lambda = 0.043 + 17.81\alpha_w - 39.85\alpha_w^2 + 36.0\alpha_w^3$ (20)

207 $\alpha_w = x_w \frac{P}{P_{sat}} + 2s$ (21)

208 **2.4 Numerical solution**

209 The numerical solution of the fully coupled governing equations was based on the FEM (finite
210 element method). First of all, mesh was developed over the computational domain by dividing it into
211 numerous elements. The distance between each element is known as the step. At each step, the
212 equations accounting for different phenomena were fully coupled and computed with the boundary
213 conditions. Initial value was given to each parameter at the first attempt then followed by an iterative
214 process until the calculation error was smaller than 10^{-5} . Commercial software COMSOL
215 Multiphysics 4.4 was used to implement the fully coupled equations. The key to successfully solving
216 this model is simulating the membrane/ionomer water content, which significantly affects many
217 critical parameters, i.e. ionomer volume fraction, EOD coefficient, ionic conductivity, hydraulic
218 permeability and water diffusivity.

219 **2.5 Mesh independence**

220 Theoretically, the computation error in the solution related to the grid must disappear for an
221 increasingly fine mesh. The effect of mesh on the final simulation results is more significant at high
222 current density due to the increasing impact of mass transport. The current density at a low cell
223 voltage (0.2 V) was therefore taken as the parameter to evaluate nine mesh grids (see [Table 5](#)) and
224 determine the influence of the element number on the solution. In this study, the number of elements
225 on various computational domains, including flow channel, GDL, CL and membrane, was
226 investigated with a fixed number of elements (250) along the channel [35]. Three levels of element
227 numbers were studied in each domain, which guaranteed the total number of element increasing at

228 1×10^4 each time. Fig. 2 shows that the current density reaches an asymptotic value while the
229 computation duration increases as the number of elements increases. The current density almost kept
230 as a constant when a finer mesh as Grid 7 was applied. A relative rapidly increase in computational
231 duration was observed from Grid 5 to Grid 7. This indicated that the time consuming step among the
232 computational process was in the catalyst layers, in which numbers of transport and electrochemical
233 processes were involved. In order to reduce the calculation error, finer mesh has to be developed on
234 catalyst layers. Having balancing both the computational accuracy and duration, Grid 6 was selected
235 considering a sufficient reliability to ensure mesh independence with acceptable calculating time.

236 **3. Results and discussions**

237 **3.1 Model validation**

238 Fig. 3 shows the comparison of the simulation results with experimental data obtained both
239 in-house and selected from literature [40]. For the in-house case shown in Fig. 3(a), the anode and
240 cathode catalyst layers were made from 20% Pt/C with a Pt loading of 0.1 and 0.4 mg cm⁻²,
241 respectively. 5% Nafion[®] ionomer solution in de-ionized water was used as binder. The ionomer mass
242 ratios were 20% in the catalyst layers at both the anode and cathode. Nafion[®] 112 membrane was used
243 as electrolyte sandwiched between anode and cathode. The graphite plates with an active area of 1.0
244 cm × 1.0 cm were grooved with serpentine gas channels with a dimension of 0.1 cm × 0.1 cm. For the
245 case in Fig. 3(b), 40% Pt/C with a platinum loading of 0.4 mg cm⁻² was used. Serpentine flow field
246 was applied as well with a dimension of the 0.1 cm × 0.1 cm and an active area of 7.2 cm × 7.2 cm.
247 For the formal case, the cell was tested with the cell and gas inlet temperatures at 80 °C under a
248 hydrogen flow rate of 200 sccm at the anode side and an air flow rate of 500 sccm at the cathode side.
249 For the latter case, cell and gas inlet temperatures were 70 °C with the hydrogen flow rate of 1200

250 sccm and air flow rate of 2200 sccm. Reactant gas pressure and humidity were kept at 1.0 atm and
251 100% for both cases. The parameters used for model validation and base case in this study are listed
252 in Table 6. The cathode transfer coefficient (α_c) was obtained by fitting the experimental polarisation
253 curves in the kinetics control zone (cell voltage higher than 0.8 V) and the obtained α_c was further
254 verified by the Tafel slope worked out in the model [39]. More details of the MEA (membrane
255 electrode assembly) preparation and cell test can be found elsewhere [36-38, 40].

256 A good agreement between the simulation results and the experimental data is indicated by Fig. 3 at
257 higher cell voltages, corresponding to lower current densities, due to the slow reaction rate and
258 insignificant mass transport impact. There was a typical drop in cell voltage due to various losses in
259 terms of activation polarization loss, ohmic loss and concentration polarization loss, as the current
260 density increased. A more rapid drop in current density, caused by the increasing mass transport
261 resistance, was observed at high current densities (mass transport control zone), where the departures
262 of simulation results compared with experimental data was clearly observed. The rapid decrease in
263 current density can be explained by the increase in mass transfer losses due to the oxygen diffuses to
264 and then adsorbs on catalyst surface to perform electrochemical reactions, which is determined by
265 both the electrode structure and the reactant provided, e.g. the mole fraction of oxygen against
266 nitrogen and water vapour in air at the cathode side. When the balance between diffusion and
267 consumption rates is achieved, a limit current density is reached. The formation of liquid water inside
268 the electrode void space restricts oxygen diffusion, especially if it is accumulated liquid water near the
269 downstream channel. However, the 2D model in this study failed to fully represent the whole MEA,
270 leading to an under-estimation of the impact of mass transfer losses in the downstream channel.
271 Especially for a large active area in the case of Wang [40], the departure of simulation result and

272 experimental data is more apparent.

273 **3.2 Comparison of different mechanisms**

274 Three models of describing the effect of liquid water generation were compared, based on the
275 polarization curves, through the reaction mechanism: traditional B-V, (1-s) corrected B-V and
276 agglomerate models. The results are shown in Fig. 4 and the channel lengths are 1 and 10 cm in Fig.
277 4(a) and Fig. 4(b), respectively. The current density predicted using (1-s) corrected B-V model was
278 slightly lower than that by traditional B-V model in the full range of cell voltage. Compared with Fig
279 3, we can see that the agglomerate model is in better agreement with the experimental results, while
280 the (1-s) corrected B-V model still shows limitations in describing the significant mass transport
281 resistance at high current density. This is because the cell performance at high current density is
282 determined by mass transport rather than electrochemical kinetics. Even through the predicted current
283 densities are deduced by considering the partial occupation of platinum active site by liquid water, the
284 (1-s) corrected B-V model fails to take the gas diffusion resistance through the ionomer film into
285 account. More rapid drop in current density was observed at high current densities in Fig. 4(b) in
286 comparison with that in Fig. 4(a), which indicated that the impact of mass transport on cell
287 performance at high current densities was more pronounced for long flow channels than for short
288 channels. This can be explained by the larger fuel concentration gradient and more liquid water
289 accumulation in the downstream channel. It is also clear, due to slow reaction rate and low mass
290 transport impact at low current densities, the results predicted by the agglomerate model were very
291 close to that of B-V model. However, the agglomerate model is capable of capturing the impact of
292 mass transport at high current densities by considering the species transport resistance through the
293 ionomer and liquid water films surrounding agglomerates.

294 **3.3 Distributions of reactant gases**

295 The mole fractions of hydrogen and oxygen within electrode and flow channel at various current
296 densities from 0.2 to 1.4 A cm⁻² are shown in Fig. 5. It is clear the concentrations of both hydrogen
297 and oxygen decrease along the diffusion direction and the concentration gradient is more apparent at
298 high current densities. Hydrogen consumption near the anode outlet is observed from the distribution
299 of hydrogen mole fraction shown in Fig. 5(a). This can be explained by the electrochemical reaction
300 and the effect of EOD (electro-osmotic drag), especially at high current densities. The increase in
301 current density from 0.6 to 1.0 A cm⁻² leads to a 69% increase in the rate of hydrogen consumption by
302 HOR and a 63% increase in the water flux under the driving force of EOD. In comparison with
303 hydrogen concentration distribution in flow channel, Fig. 5(b) indicates that, oxygen remains at a very
304 high concentration close to the inlet level in flow channel, in the full range of current densities from
305 0.2 to 1.4 A cm⁻², along the air flow direction. On the contrary, a significant concentration gradient is
306 shown through the diffusion direction as the current density increases, which is in consistence with
307 the previous modelling results of Nguyen et al. [41]. This can be explained by the increase in oxygen
308 consumption due to the accelerated ORR and the presence of large oxygen transport resistance
309 through the electrode. In addition, the more water generated at high current density dilutes the oxygen.
310 The formation of liquid water reduces the porosity of the electrode and increases the thickness of
311 liquid water film surrounding the agglomerate, leading to a further decrease in the oxygen diffusivity
312 through the electrode before it can reach the active sites. However this does not happen at the anode
313 side due to the much easier diffusion of the smaller hydrogen molecular and its relative high mole
314 fraction.

315 **3.4 Distributions of effectiveness factor**

316 The effectiveness factors, used to evaluate how effective the catalyst layer is utilized, of the ACL
317 and CCL at various current densities from 0.3 to 1.2 A cm⁻² are shown in Fig. 6. In Fig. 6(a), X=0 and
318 X=1 represent the interfaces of GDL-CL and CL-membrane of anode, while Y=0 and Y=1 represent
319 the anode inlet and outlet, respectively. In Fig. 6(b), X=0 and X=1 represent the interfaces of
320 membrane-CL and CL-GDL of cathode, while Y=0 and Y=1 represent the cathode outlet and inlet,
321 respectively. The highest effectiveness factors, for both the ACL and CCL, are observed at 0.3 A cm⁻²,
322 which decrease with the increase of the current density to 1.2 A cm⁻². The decrease in effectiveness
323 factor can be explained by the increased electrochemical reaction rate as the current density increases.
324 At this stage, the overall reaction rate is mainly determined by the rate of chemical reaction rather
325 than mass transport. As the current density increases, the increase in electrochemical reaction rate
326 leads to a faster consumption of reactant gases relative to the diffusion rate to the active sites. The rate
327 determining process therefore changes from the electrochemical reaction to the reactant gas diffusion.
328 Consequently, along the gas diffusion direction through catalyst layers, the catalyst utilization is
329 higher near the GDL-CL interface (X=0 for anode and X=1 for cathode) than that of the CL-membrane
330 interface (X=1 for anode and X=0 for cathode), which agree with the finding of Sun et al. [25]. It is
331 important to note that the change of the effectiveness factor is not pronounced along the reactant gas
332 flow direction (Y direction). This is because the gases are supplied above stoichiometric requirements.
333 Reactant gases are provided at such rates to guarantee the almost uniform concentrations along the
334 channels. A comparison of Fig. 6(a) and Fig. 6(b) shows a much higher effectiveness factor of ACL
335 than that of CCL, especially at high current densities. The ACL effectiveness factor remains high, ca.
336 80%, for a certain distance along the hydrogen diffusion. On the contrary, due to the large oxygen
337 transport resistance through the cathode, the effectiveness factor of CCL is much lower compared

338 with that of ACL. The platinum catalyst in the CCL located near the CL-GDL interface cannot be
339 fully utilized, resulting in a waste of expensive catalyst.

340 **3.5 Distributions of current density**

341 [Fig. 7](#) shows the current density distributions within the catalyst layers of anode and cathode at
342 various cell voltages from 0.7 to 0.4 V. The current densities increase in both catalyst layers as the cell
343 voltage decreases. It can be seen that the current densities decrease along the reactant gas diffusion
344 direction at both the anode and cathode, leading to insufficient utilization of the catalyst near the
345 CL-membrane boundary ($X=0$ for cathode and $X=1$ for anode). On the contrary, the current densities
346 almost remain constant along the reactant flow direction. It is apparent that the current density
347 distribution in the ACL is more uniform than that in the CCL. In the region near the CL-membrane
348 interface, the anode current density is higher than the cathode current density at a fixed cell voltage.
349 This can be explained by the sluggish ORR in the cathode. Due to the slower ORR in the cathode, the
350 interior of the cathode catalyst layer is not fully utilised, especially at a high current density.

351 **3.6 Distributions of liquid water saturation**

352 The liquid water saturations in both electrodes, including CLs and GDLs, are shown in [Fig. 8](#). The
353 coordinates of X and Y indicate the same geometry as those in [Fig. 6](#). It is clear that the liquid water
354 saturation increases in both electrodes as the current density increases. Also, the liquid water
355 saturation is relatively high near the outlets in both electrodes, indicating more severe water flooding
356 in these areas. This finding is in consistence with the experimental measurement of Iranzo et al. [[19](#)].
357 Comparison of [Fig. 8\(a\)](#) and [Fig. 8\(b\)](#) shows that the gradient of liquid water saturation is greater in
358 cathode electrode than of the anode, especially along the channel direction.

359 **3.7 Distributions of temperature**

360 The temperature profiles at various current densities from 0.6 to 1.0 A cm⁻² are shown in Fig. 9. A
361 non-uniform temperature distribution is clearly indicated. The temperature increases along the air
362 flow and decreases from the cathode to the anode through the membrane with the highest value at the
363 cathode side. The most significant temperature rise, as expected, is observed in the cathode, leading to
364 a ca. 4 °C difference between anode and cathode at 1.0 A cm⁻². This temperature increase at the
365 cathode can be explained by both the exothermic ORR and water phase-transfer occurred. The latter
366 comprises ca. 6% of the total heat released at 1.0 A cm⁻² (Table 7), which is the latent heat associated
367 with water phase change from dissolved water to liquid water during membrane/ionomer desorption.
368 A temperature rise along the cathode flow channel is clearly shown in Fig. 9. This can be explained by
369 the heat released by water phase-transfer as liquid water is pushed downstream to the channel exit by
370 the airflow.

371 As a main heat source, the heat released by ORR comprises more than 90% of the total heat
372 released at 1.0 A cm⁻². Similar to the ORR in the cathode, the HOR in the anode occupies more than
373 88% of the total heat change, which therefore can be considered as the main heat source in the anode.
374 Due to the endothermic nature and large contribution of the HOR to the total heat in the anode, the
375 anode is endothermic in full range of current densities. With the increase in current density, the ratio
376 of HOR to the total heat in the anode increases whereas that of ORR to the total heat in the cathode
377 decreases. In the anode, the heat released by ionomer water uptake is the second main heat source,
378 which comprises ca. 10% of the total heat in the anode at 0.2 A cm⁻². As the current density increases,
379 there is a decrease in the contribution of the heat released by ionomer water uptake in the anode. This
380 is caused by a relative fast increase in the heat absorption due to the HOR. Although the absolute heat
381 released by ionomer water uptake increases, the ratio decreases from ca. 11% to 3% as the current

382 density increases from 0.2 to 1.0 A cm⁻². Table 7 also shows that the heat contribution of ORR
383 decreases and ionomer desorption increases with the increase in current density in the cathode. This is
384 because more dissolved water is generated at a high current density, which then transfers to liquid
385 water by ionomer desorption after the ionomer is fully saturated.

386 The detailed temperature profiles at the anode channel-GDL interface are shown in Fig. 10. The
387 temperature increases sharply along the hydrogen flow direction until a maximum temperature is
388 achieved, then slightly decreases near the anode outlet. The maximum temperature is located in the
389 middle of the channel near to the inlet. The temperature rise in the anode is much less than that of the
390 cathode due to the endothermic HOR within the anode catalyst layer (Fig. 9). This increase can be
391 explained by the heat transferred from the cathode and the latent heat released via water phase-transfer.
392 As shown in Table 7, the heat via water phase-transfer approximately comprises 11% of total heat in
393 the anode at 0.2 A cm⁻², which decreases to less than 3% as the current density increases to 1.0 A cm⁻².
394 Therefore, heat transfer from the cathode is the main heat source contributed to the temperature rise in
395 the anode.

396 **3.8 Comparison of the isothermal and non-isothermal model**

397 Fig. 11 shows the liquid water saturation at the cathode CL-GDL interface at 1.0 A cm⁻² simulated
398 by both the isothermal and non-isothermal models. The dimensionless coordinate is the same as in Fig.
399 1. The liquid water saturation increases along the air flowing direction at the cathode GDL-CL
400 interface. By taking the temperature rise into account, the liquid water saturation predicted by the
401 non-isothermal model is slightly smaller than that by the isothermal model, caused by the fast increase
402 in the saturation pressure of water vapour (p_{sat}) with temperature. Specifically, p_{sat} increases from
403 31.16 to 36.99 kPa as temperature increases from 70 to 74 °C. The increase in the saturation pressure

404 of water vapour, improves the WCC (water carrying capacity) of gas mixture, thus to a certain extent,
405 prohibits the water phase-transfer from vapour to liquid water. Furthermore, the temperature rise is
406 more apparent at a high operating temperature in comparison with a low one [37]. The heat released
407 during fuel cell operation can help mitigate water flooding by improving the WCC of the reactant
408 gases. However, this is at the expense of reducing the effective oxygen concentration of the cathode
409 reactant gas, which may results in a more significant decline in cell performance than that induced by
410 flooding. The mole fraction of oxygen in a fully humidified mixture decreases from 18.24% to 13.34%
411 as temperature increases from 70 to 74 °C, which may lead to oxygen starvation and damage to the
412 cathode. Thus, for a fuel cell operated at a high temperature, the performance may be limited by the
413 excessive moisture in the cathode reactant gas.

414 **3.9 Regressed expressions for liquid water saturation**

415 Fig. 12 shows the liquid water saturation in both electrodes (including CLs and GDLs) at various
416 current densities. It indicates that the increase of the current density leads to a logarithmic increase in
417 the liquid water saturation, which is greater in the cathode than that in the anode. In addition, the
418 liquid water saturations are slightly larger in CLs than that in GDLs at both electrodes, especially in
419 the cathode due to the water generated via ORR.

420 Mathematical expressions are regressed in terms of logarithmic and exponential equations to fit the
421 average liquid water saturation in both electrodes, as shown in Fig. 13, respectively. Linear
422 logarithmic transform function (Log3P1) and two-phase exponential decay function with time offset
423 (ExpDecay2) were selected as regression functions. The cathode average liquid water saturation (s_c) in
424 Fig. 13(a) could be well fitted by a logarithmic expression (Eq. 22) with a R^2 coefficient of 99.33%.
425 (R^2 is a statistical measure of how close the data to the fitted regression line). However, a large error

426 of fitting occurs in medium and high current densities when applying the same regression function to
 427 the anode average liquid water saturation (s_a). The logarithmic expression (Eq. 23) led to a R^2
 428 coefficient of only 94.26% and a remarkable standard error of parameter c . Thus, the logarithmic
 429 expression of $s_a - i$ was considered as an unconfident relationship without enough accuracy. Instead,
 430 an exponential expression (Eq. 24), more complex in form in comparison with logarithmic expression,
 431 is regressed in Fig. 13(b). It can be seen that the exponential express is more accurate here.

$$432 \quad s_c = 0.1344 + 0.05912 \ln(i + 0.15109) \quad (R^2 > 99.33\%) \quad (22)$$

$$433 \quad s_a = 0.07405 + 0.01354 \ln(i + 3.00567 \times 10^{-4}) \quad (R^2 > 94.26\%) \quad (23)$$

$$434 \quad s_a = 0.10232 - 0.06965 \exp\left(-\frac{i + 0.00489}{1.12255}\right) - 0.0378 \exp\left(-\frac{i + 0.00489}{0.02805}\right) \quad (R^2 > 99.88) \quad (24)$$

435 The average liquid water saturations in both electrodes with respect to the current density higher
 436 than 0.6 A cm^{-2} are shown in Fig. 14. Log3P1 function, which is used to fit the data plotted, results in
 437 accurate expressions (Eq. 25 and Eq. 26) with R^2 greater than 99.9% in both electrodes. This indicates
 438 that the changes in the average liquid water saturation in both electrodes follows a logarithmic
 439 relationship at current densities higher than 0.6 A cm^{-2} . Comparing Fig. 14 with Fig. 13(b) suggests
 440 that, as the current density increases, the increase in the average liquid water saturation in the anode
 441 firstly obeys exponential function at low current densities followed by logarithmic function at high
 442 current densities. However, it is insufficient to describe the relationship between the anode average
 443 liquid water saturation and current density using a logarithmic function in full range of current
 444 densities.

$$445 \quad s_c = 0.11463 + 0.07763 \ln(i + 0.40119) \quad (R^2 > 99.99\%) \quad (25)$$

$$446 \quad s_a = 0.06909 + 0.02897 \ln(i + 0.16568) \quad (R^2 > 99.90\%) \quad (26)$$

447 The relationship between average liquid water saturation in the anode and the cathode is regressed

448 in Fig. 15. The data plotted can be roughly divided into three segments, corresponding to the liquid
 449 water saturation at the low, medium and high levels. In order to guarantee the accuracy, the R^2
 450 coefficient of each regression expression within each segment is controlled greater than 99.9%. The
 451 results indicate that at the low and high liquid water saturation, a linear relationship exists between the
 452 average liquid water saturations in the anode and the cathode. However, at the medium liquid water
 453 saturation, the exponential function is more accurate than the linear relationship. The relation of liquid
 454 water saturation at the anode and cathode is similar to the modelling results of Jiang and Wang [42].

455 As stated by O'Rourke et al. [20], the water flooding in anode could be detected prior to severe
 456 damage of cathode catalyst, which could be avoided with the aid of the regressed expressions of the
 457 liquid water saturation in the anode and the cathode.

$$458 \quad s_c = 3.65764s_a - 1.45542 \times 10^{-4} \quad (R^2 > 99.99\%) \quad \text{low water saturation} \quad (27)$$

$$459 \quad s_c = \exp(27.0888s_a^2 + 31.24035s_a - 4.00174) \quad (R^2 > 99.44\%) \quad \text{medium water saturation} \quad (28)$$

$$460 \quad s_c = 2.32873s_a - 0.03044 \quad (R^2 > 99.96\%) \quad \text{high water saturation} \quad (29)$$

461 **4. Conclusion**

462 A two-dimensional, two-phase flow, along-the-channel, non-isothermal, CFD model for a single
 463 PEMFC is developed. Water was treated as three different phases: vapour, dissolved and liquid water.
 464 The water phase-transfer between each phase, associated with the combinational transport mechanism
 465 through the membrane, is numerically studied. Three types of models are compared for hydrogen
 466 reduction and oxygen oxidation in catalyst layers, including traditional B-V, (1-s) corrected B-V and
 467 agglomerate mechanism. Temperature changes in MEA and channels due to electrochemical reaction,
 468 ohmic resistance and water phase change are analysed and the contribution of each heat source is
 469 summarised and compared at a variety of current densities. Mathematical expressions for liquid water

470 saturations against current density at both anode and cathode are regressed and the mathematical
471 relationship between them is developed. Analysis of the simulation results leads to the following
472 insights:

473 The current density predicted by traditional B-V, (1-s) corrected B-V and agglomerate models are
474 in general agreement with experimental data at low current densities. As the current density increases,
475 by accounting for the extra oxygen diffusion resistance through the ionomer/water film and the loss of
476 catalyst layer porosity due to ionomer swelling, the agglomerate model gives more accurate
477 simulation results in comparison with the others. However, due to significant water flooding in
478 downstream channel, the 2D model developed cannot fit the experimental data very well at high
479 current densities, especially when long flow channels are applied. Due to the more significant
480 transport resistance of oxygen in the cathode than that of hydrogen in the anode, the effectiveness
481 factor of the cathode catalyst layer is much lower than that of the anode catalyst layer.

482 Water flooding, represented by the liquid water saturation, is prone to occur near the downstream
483 channel of both the anode and cathode. Liquid water saturation is temperature dependent, which
484 requires the non-uniform distribution of temperature predicted by the non-isothermal model. The most
485 significant temperature rise is in the cathode catalyst layer due to the contribution of exothermic ORR,
486 which comprises more than 90% of total heat released. With the increase in the current density, the
487 heat contribution of ORR decreases while ionomer/membrane desorption increases. On the contrary,
488 the endothermic HOR is the main heat source in the anode. At a low current density, the heat released
489 by ionomer/membrane water uptake contributes ca. 10% to the total heat in the anode.

490 The mathematical expressions for the relationship between the current density and liquid water
491 saturations in both the anode and the cathode are regressed. Nonlinear current density – water

492 saturation relations are found at both electrodes. It is believed that the liquid water saturation in the
493 cathode presents logarithm relevant to current density. However, the liquid water saturation in the
494 anode increases exponentially as the current density increase. At a high current density greater than
495 0.6 A cm^{-2} , the liquid water saturations at both electrodes can be described by logarithmic relationship.
496 The relationship between liquid water saturations in the anode and the cathode is also regressed. It is
497 found that at low and high current densities, the relationship can be accurately represented by linear
498 function, but exponential function is believed to be more accurate at medium current densities.

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503 **References**

- 504 [1] Gottesfeld S. Fuel cell techno-personal milestones 1984-2006. *J Power Sources* 2007;171:37-45.
- 505 [2] Dai W, Wang H, Yuan XZ, Martin JJ, Yang D, Qiao J, Ma J. A review on water balance in the
506 membrane electrode assembly of proton exchange membrane fuel cells. *Int J Hydrogen Energy*
507 2009;34:9461-78.
- 508 [3] Wang Y, Chen KS, Mishler J, Cho SC, Adroher XC. A review of polymer electrolyte membrane
509 fuel cells: Technology, applications, and needs on fundamental research. *App Energy*
510 2011;88:981-1007.
- 511 [4] Wang J. Barriers of scaling-up fuel cells: Cost, durability and reliability. *Energy* 2015;80:509-21.
- 512 [5] Lund H, Andersen AN, Østergaard PA, Mathiesen BV, Connolly D. From electricity smart grids
513 to smart energy systems – A market operation based approach and understanding. *Energy*
514 2012;42:96-102.
- 515 [6] Rokni M. Thermodynamic and thermoeconomic analysis of a system with biomass gasification,

516 solid oxide fuel cell (SOFC) and Stirling engine. *Energy* 2014;76:19-31.

517 [7] Cappa F, Facci AL, Ubertini S. Proton exchange membrane fuel cell for cooperating households:
518 A convenient combined heat and power solution for residential applications. *Energy*
519 2015;90:1229-38.

520 [8] Carton JG, Olabi AG. Wind/hydrogen hybrid systems: Opportunity for Ireland's wind resource to
521 provide consistent sustainable energy supply. *Energy* 2010;35:4536-44.

522 [9] Djilali N. Computational modelling of polymer electrolyte membrane (PEM) fuel cells:
523 Challenges and opportunities. *Energy* 2007;32: 269-80.

524 [10] Bazylak A. Liquid water visualization in PEM fuel cells: A review. *Int J Hydrogen Energy*
525 2009;34:3845-57.

526 [11] Carton JG, Lawlor V, Olabi AG, Hochenauer C, Zauner G. Water droplet accumulation and
527 motion in PEM (Proton Exchange Membrane) fuel cell mini-channels. *Energy* 2012;39:63-73.

528 [12] Ge S, Li X, Yi B, Hsing IM. Absorption, desorption, and transport of water in polymer electrolyte
529 membranes for fuel cells. *J Electrochem Soc* 2005;152:A1149-57.

530 [13] Wu H, Berg P, Li X. Steady and unsteady 3D non-isothermal modeling of PEM fuel cells with
531 the effect of non-equilibrium phase transfer. *App Energy* 2010;87:2778-84.

532 [14] Yang XG, Ye Q, Cheng P. Matching of water and temperature fields in proton exchange
533 membrane fuel cells with non-uniform distributions. *Int J Hydrogen Energy* 2011;36:12524-37.

534 [15] Das PK, Li X, Liu ZS. Analysis of liquid water transport in cathode catalyst layer of PEM fuel
535 cells. *Int J Hydrogen Energy* 2010;35:2403-16.

536 [16] Meng H, Wang CY. Model of Two-Phase Flow and Flooding Dynamics in Polymer Electrolyte
537 Fuel Cells. *J Electrochem Soc* 2005;152:A1733-41.

538 [17] Ferreira RB, Falcao DS, Oliveira VB, Pinto AMFR. Numerical simulation of two-phase flow in
539 an anode gas channel of a proton exchange membrane fuel cell. *Energy* 2015;82:619-28.

540 [18] Wong KH, Loo KH, Lai YM, Tan SC, Tse CK. A theoretical study of inlet relative humidity
541 control in PEM fuel cell. *Int J Hydrogen Energy* 2011;36:11871-85.

- 542 [19] Iranzo A, Boillat P, Biesdorf J, Salva A. Investigation of the liquid water distribution in a 50 cm²
543 PEM fuel cell: Effects of reactants relative humidity, current density, and cathode stoichiometry.
544 Energy 2015;82:914-21.
- 545 [20] O'Rourke J, Ramani M, Arcak M. In situ detection of anode flooding of a PEM fuel cell. Int J
546 Hydrogen Energy 2009;34:6765-70.
- 547 [21] Anderson R, Blanco M, Bi X, Wilkinson DP. Anode water removal and cathode gas diffusion
548 layer flooding in a proton exchange membrane fuel cell. Int J Hydrogen Energy 2012;37:
549 16093-103.
- 550 [22] Wang ZH, Wang CY, Chen KS. Two-phase flow and transport in the air cathode of proton
551 exchange membrane fuel cells. J Power Sources 2001;94:40-50.
- 552 [23] He G, Ming P, Zhao Z, Abudula A, Xiao Y. A two-fluid model for two-phase flow in PEMFCs. J
553 Power Sources 2007;163:864-73.
- 554 [24] Ferreira RB, Falcao DS, Oliveira VB, Pinto AMFR. A one-dimensional and two-phase flow
555 model of a proton exchange membrane fuel cell. J Chem Technol Biotechnol 2015; 90:1547-51.
- 556 [25] Sun W, Peppley BA, Karan K. An improved two-dimensional agglomerate cathode model to
557 study the influence of catalyst layer structural parameters. Electrochim Acta 2005;50:3359-74.
- 558 [26] Shah AA, Kim GS, Sui PC, Harvey D. Transient non-isothermal model of a polymer electrolyte
559 fuel cell. J Power Sources 2007;163:793-806.
- 560 [27] Ju H, Meng H, Wang CY. A single-phase, non-isothermal model for PEM fuel cells. International
561 J Heat and Mass Transfer 2005;48:1303-15.
- 562 [28] Wang MH, Guo H, Ma CF. Temperature distribution on MEA surface of a PEMFC with
563 serpentine channel flow bed. J Power Sources 2006;157:181-7.
- 564 [29] Zhang GS, Guo LJ, Ma LZ, Liu HT. Simultaneous measurement of current and temperature
565 distributions in a proton exchange membrane fuel cell. J Power Sources 2010;195:3597-604.
- 566 [30] Lin H, Cao TF, Chen L, He YL, Tao WQ. In situ measurement of temperature distribution within
567 a single polymer electrolyte membrane fuel cell. Int J Hydrogen Energy 2012;37:11871-86.

- 568 [31] Pasaogullari U, Wang CY. Two-phase transport and the role of microporous layer in polymer
569 electrolyte fuel cells. *Electrochim Acta* 2004;49:4359-69.
- 570 [32] Wu H, Berg P, Li X. Modelling of PEMFC transients with finite-rate phase transfer processes. *J*
571 *Electrochem Soc* 2010;157:B1-12.
- 572 [33] Wang CY, Groll M, Rosler JS, Tu CJ. Porous medium model for two-phase flow in mini channels
573 with applications to micro heat pipes. *Heat Recovery Syst CHP* 1994;14:377-90.
- 574 [34] Xing L, Mamlouk M, Scott K. A two dimensional agglomerate model for a proton exchange
575 membrane fuel cell. *Energy*, 2013;61:196-210.
- 576 [35] Xing L, Song X, Scott K, Picker V, Cao W. Multi-variable optimization of PEMFC cathodes
577 based on surrogate modelling. *Int J Hydrogen Energy* 2013;38:14295-313.
- 578 [36] Xing L, Mamlouk M, Kumar R, Scott K. Numerical investigation of the optimal Nafion®
579 ionomer content in cathode catalyst layer: An agglomerate two-phase flow modelling. *Int J*
580 *Hydrogen Energy* 2014;39:9087-104.
- 581 [37] Xing L, Liu X, Alaje T, Kumar R, Mamlouk M, Scott K. A two-phase flow and non-isothermal
582 agglomerate model for a proton exchange membrane (PEM) fuel cell. *Energy* 2014;73:618-34.
- 583 [38] Xing L, Das PK, Song X, Mamlouk M, Scott K. Numerical analysis of the optimum
584 membrane/ionomer water content of PEMFCs: The interaction of Nafion® ionomer content and
585 cathode relative humidity. *Appl Energy* 2014;138:242-57.
- 586 [39] Barbir F. *PEM fuel cells: theory and practice*. 1st ed. Oxford: Elsevier Academic Press; 2005.
- 587 [40] Wang L, Husar A, Zhou T, Liu H. A parametric study of PEM fuel cell performances. *Int J*
588 *Hydrogen Energy* 2003;28:1263-72.
- 589 [41] Nguyen PT, Berning T, Djilali N. Computational model of a PEM fuel cell with serpentine gas
590 flow channels. *J. Power Sources* 2004;130:149-57.
- 591 [42] Jiang F, Wang C-Y. Numerical modeling of liquid water motion in a polymer electrolyte fuel cell.
592 *Int J Hydrogen Energy* 2014;39:942-50.

593

594 **Nomenclature**

A_s	reaction surface area per unit platinum mass, $\text{m}^2 \text{kg}^{-1}$
a	specific area, m^{-1}
$c_{p,i}$	specific heat capacity of species i , $\text{J mol}^{-1} \text{K}^{-1}$
c	concentration, mol m^{-3}
D	diffusivity, $\text{m}^2 \text{s}^{-1}$
D_c	capillary diffusion coefficient, $\text{m}^2 \text{s}^{-1}$
D_{ij}	Maxwell-Stefan diffusion coefficient matrix, $\text{m}^2 \text{s}^{-1}$
E	effectiveness factor
E^0	open circle potential, V
E_{cell}	cell voltage, V
EW	equivalent weight of Nafion [®] membrane, g mol^{-1}
F	Farady's constant, 96485 C mol^{-1}
f	platinum mass ratio to Pt/C
H	Henry's constant, $\text{Pa m}^3 \text{mol}^{-1}$
i	current density, A m^{-2}
i_0	exchange current density, A m^{-2}
$J(s)$	Leverett function
k	rate coefficient, s^{-1}
k_i	Thermal conductivity of species i , $\text{W m}^{-1} \text{K}^{-1}$
k_r	relative permeability
k_p	hydraulic permeability, m^2

l	thickness, m
L	volume fraction
M_j	molecular weight for specie j, kg mol ⁻¹
M_T	Thieles's modulus
m	mass loading, mg cm ⁻²
N	number per volume, m ⁻³
n	number
p	pressure, Pa
R	ideal gas constant, 8.314 J mol ⁻¹ K ⁻¹
R_M	membrane resistance, Ω m ⁻²
RH	relative humidity
r	radius, m
S	source term
s	liquid water saturation
T	temperature, K
\mathbf{u}	velocity vector, m s ⁻¹
V	mole volume, m ³
w	mass fraction
X	normalised distance (x/δ_{CL})
x	mole fraction
Y	normalised distance ($z/(z_{CC+} + z_{Ch})$)
$\%M$	volume fraction of primary pores occupied by ionomer

595

596 *Greek*

α	charge transfer coefficient
α_w	water activity
λ	water content
μ	viscosity, Pa s
ρ	density, kg m ⁻³
ε	porosity
δ	thickness of ionomer/liquid water coating, m
γ	oxygen diffusion rate through the coating, s ⁻¹
η	overpotential, V
σ	surface tension, N m ⁻¹
θ_c	contact angel, °
σ_s	electronic conductivity, S m ⁻¹
σ_M	ionic conductivity, S m ⁻¹
φ	potential, V
Ψ_w	association parameter for water (the value is 2.6)

597 *Superscripts*

<i>0</i>	intrinsic
<i>d</i>	dissolved
<i>eff</i>	effective
<i>ref</i>	reference

	<i>eq</i>	equilibrium
	<i>l</i>	liquid
	<i>g</i>	gas
598	<i>Subscripts</i>	
	<i>a</i>	anode
	<i>ads</i>	adsorption
	<i>agg</i>	agglomerate
	<i>C</i>	carbon
	<i>c</i>	cathode
	<i>CL</i>	catalyst layer
	<i>des</i>	desorption
	<i>GDL</i>	gas diffusion layer
	<i>i</i>	species i
	<i>j</i>	species j
	<i>Kn</i>	Knudsen diffusion
	<i>M</i>	Membrane/ionomer
	<i>P</i>	void space
	<i>Pt</i>	platinum
	<i>Pt/C</i>	platinum dispersed carbon
	<i>p</i>	primary pores
	<i>r</i>	relative
	<i>S</i>	GDL penetration

<i>s</i>	secondary pores
<i>T</i>	temperature
<i>w</i>	liquid water
<i>sat</i>	saturation
<i>tot</i>	total
<i>vl</i>	vapour to liquid
<i>vd</i>	vapour to dissolved
<i>dl</i>	dissolved to liquid