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Anode partial flooding modelling of proton exchange membrane fuel cells: Optimisation of electrode properties and channel geometries

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HIGHLIGHTS

- A fully coupled 2D, along-the-channel, two-phase flow, non-isothermal, CFD model is developed.
- More severe water flooding at the cathode than that at the anode is numerically demonstrated.
- Effects of electrode properties and channel geometries on fuel cell performance are studied.
- Thinner GDL could result in more non-uniform and more significant temperature rise at high current densities.
- Novel channel design featured with multi-inlets and outlets is proposed to reduce water flooding.

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ABSTRACT

A two-dimensional, along-the-channel, two-phase flow, non-isothermal model is developed which represents a low temperature proton exchange membrane (PEM) fuel cell. The model describes the liquid water profiles and heat distributions inside the membrane electrode assembly (MEA) and gas flow channels as well as effectiveness factors of the catalyst layers. All the major transport and electrochemical processes are taken into account except for reactant species crossover through the membrane. The catalyst layers are treated as spherical agglomerates with inter-void spaces, which are in turn covered by ionomer and liquid water films. Liquid water formation and transport at the anode is included while water phase-transfer between vapour, dissolved water and liquid water associated with membrane/ionomer water uptake, desorption and condensation/evaporation are considered. The model is validated by experimental data and used to numerically study the effects of electrode properties (contact angel, porosity, thickness and platinum loading) and channel geometries (length and depth) on liquid water profiles and cell performance. Results reveal low liquid water saturation with large contact angle, low electrode porosity and platinum loading, and short and deep channel. An optimal channel length of 1 cm was found to maximise the current densities at low cell voltages. A novel channel design featured with multi-outlets and inlets along the channel was proposed to mitigate the effect of water flooding and improve the cell performance.

1. Introduction

Proton exchange membrane (PEM) fuel cells are considered as promising candidate in automotive, portable and residential applications benefited from their high efficiency, high volumetric power density and zero emission of greenhouse gases during operation (Wang et al., 2011; Lund et al., 2012; Gahleitner, 2013). Many challenges are faced for large scale PEMFC commercialisation, and water management is considered as a critical one, especially for the low/medium temperature PEMFCs. Water is unavoidable in PEMFCs due to the oxygen reduction reaction (ORR) at the cathode and the use of humidified reactant gases at both cathode and anode. An adequate amount of water is essential to a successful fuel cell operation, because a hydrated membrane and ionomer in the catalyst layers is required to maintain high conductivity. However, excess water may cause flooding inside the

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porous electrode and channel, resulting in a pronounced decrease in mass transport rate and thus reduction in the overall fuel cell performance.

Water transport through the membrane, in terms of three mechanisms: electro-osmotic drag (EOD), back diffusion and hydraulic permeation, determines the membrane/ionomer water content, which then significantly affects the membrane/ionomer conductivity. The liquid water formed in the porous electrode is mainly attributed to membrane/ionomer desorption (the membrane/ionomer water content reaches equilibrium) and water vapour condensation (partial pressure of water vapour exceeds the saturation pressure). At the anode side, hydrogen consumption could result in the humidified anode reactant gas becoming supersaturated with water vapour, which transfers to liquid water by condensation. Experimental studies have detected liquid water in the anode; more liquid water was observed at a high anode relative humidity (Wong et al., 2011; Iranzo et al., 2015). However, anode flooding has not been considered in the majority of previous modelling research.

The amount of liquid water in the porous electrode is a consequence of the interaction between many parameters, such as contact angle, electrode porosity and microstructure, flow patterns, flow filed design, and operating conditions. Intensive experimental studies have been focused on novel gas diffusion layer (GDL) and catalyst layer (CL) materials (Park et al., 2012; Holdcroft, 2014), optimal PTFE content (Shimpalee et al., 2007; Park et al., 2008) and optimised operating conditions (Ge and Wang, 2007; Barelli et al., 2011). For a CL with a fixed thickness, the increase in platinum loading and Pt/C ratio could increase the CL porosity and the electrochemical active surface area (ECSA) (Xing et al., 2013b). The increased CL porosity facilitates gas transport and increases the CL ability to remove water due to the increased capillary diffusion coefficient (Wu et al., 2010a, 2010b; Yang et al., 2011). In the meanwhile, the increased ECSA accelerates formation of liquid water due to the faster ORR process. The role of the GDL is to facilitate the transport of reactant and product gases, liquid water and heat between the channel and CL. Reactant gases can be uniformly dispersed when diffusing through GDL before reaching catalyst sites, which can then avoid local hot spots at electrodes near the inlet, created by the non-uniform distributed reactant concentration along its flow direction. A certain thickness of GDL is required for well dispersed reactant gases but it is at the expense of higher diffusion resistance, which could limit the fuel cell performance at high current densities. For liquid water transport through the GDL, the use of higher amount of hydrophobic materials could lead to the lower level of flooding (Pasaogullari and Wang, 2004a). Application of micro-porous layers (MPL) has been an effective approach to mitigate liquid water flooding in the porous electrodes in many commercial cells. Cho and Mench (2012) directly visualised the liquid water profiles within a cell using hydrogen–deuterium contrast neutron radiography (NR), the MPL has proved to mitigate flooding in the cathode side through enhanced transport of water to the anode side. Despite many advanced understandings made by the studies, a complete investigation is still required to fully understand the role of all the factors that affect the anode performance and flooding. For example, up to now, the GDL thickness on temperature rise and strategies to mitigate flooding using novel flow field designs are still not well investigated in the literature.

Mathematical models present a good platform for the complete investigation of all the interacting parameters in fuel cells. To accurately model the physical and chemical processes in GDLs, CLs and flow channels, the two-phase flow phenomenon between liquid water and gas mixture must be carefully considered. For this, the multi-phase mixture (M^2) model was firstly developed by Wang and Cheng (1996), in which the water–gas mixture was considered as a continuum flowing in porous media under capillary force. Later on, Wang and his co-workers (Wang et al., 2001; Basu et al., 2009) combined the volume of fluid (VOF) method with the M^2 model to describe the liquid water transport in porous electrodes and flow channels. Pasaogullari and Wang (2004b) investigated the role of MPL in liquid water control using a two-phase flow model. Weber and Newman (2005) quantified the effects of MPL using a two-phase flow and robust membrane model. By considering the partial occupation of the electrode void space by liquid water, the reduction in effective mass transport coefficient was highlighted by Nam and Kaviany (2003). The usefulness of the agglomerate model for treating the two-phase flow phenomenon in PEMFCs was first demonstrated by Shah et al. (2007), in which the additional oxygen transport resistance through a thin liquid water film surrounding the agglomerate was taken into account. The agglomerate model has later been applied to the phosphoric acid doped polybenzimidazole (PBI) membrane fuel cell by Sousa et al. (2010), in which the effect of phosphoric acid loading on the cell performance was successfully studied.

Building on these studies, we have developed an anode partial flooding model (Xing et al., 2016) for PEM fuel cells. The model is a two-dimensional, along-the-channel, two-phase flow and non-isothermal model, based on a spherical-agglomerate catalyst structure, and combined with a comprehensive water phase-transfer and transport mechanism. The non-uniform distribution of membrane/ionomer water content due to the driving force of EOD, back diffusion and hydraulic permeation, the ionomer film swelling due to ionomer water uptake, as well as the water phase-change between vapour, dissolved water and liquid water are all considered. In our previous work (Xing et al., 2016), this anode partial flooding model was validated by comparing with experimental measurement. In this paper, we are using the anode partial flooding model to investigate the electrode properties, e.g. contact angle, porosity, platinum loading and GDL thickness, as well as channel geometries including length and depth, in relation to temperature distribution, water flooding and fuel cell performance. The model is also used to optimise electrode parameters and provide theoretical guidance to a novel channel design featured with multi-outlets and inlets, which is for the first time proposed.

2. Model development

2.1. Model features and assumptions

A 2D along-the-channel model for a typical fuel cell was developed. The model features and main assumptions are listed as follows:

(1) *Fuel cell geometry and computational domain.* The 2D representation of the geometry and computational domain is given in Fig. 1, showing flow channels (Domain 1), GDLs (Domain 2), and CLs (Domain 3) of both the anode and cathode, and a Nafion® membrane (Domain 4) in between. The initial input structural parameters and material properties of each layer are listed in Table 1.

(2) *Structure of the catalyst layers.* The catalyst layers are treated as spherical catalyst agglomerates with porous inter-agglomerate spaces defined as the *primary pores*. The void space between the agglomerates is defined as the *secondary pores*. As shown in Fig. 2, the *primary pores* inside the agglomerate are partially filled with the ionomer, whereas the catalyst agglomerates are in turn surrounded by ionomer and liquid water films, which partially occupy the *secondary pores* between the agglomerates.