



Numerical investigation of the effect of cathode catalyst layer structure and composition on polymer electrolyte membrane fuel cell performance

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ABSTRACT

The effect of the cathode catalyst layer's structure and composition on the overall performance of a polymer electrolyte membrane fuel cell (PEMFC) is investigated numerically. The starting point of the sub-grid scale catalyst layer model is the well-known flooded agglomerate concept. The proposed model addresses the effects of ionomer (Nafion) loading, catalyst (platinum) loading, platinum/carbon ratio, agglomerate size and cathode layer thickness. The sub-grid scale model is first validated against experimental data and previously published results, and then embedded within a two-dimensional validated computational fluid dynamics code that can predict the overall performance of the fuel cell. The integrated model is then used to explore a wide range of the compositional and structural parameter space, mentioned earlier. In each case, the model is able to correctly predict the trends observed by past experimental studies. It is found that the performance trends are often different at intermediate versus high current densities—the former being governed by agglomerate-scale (or local) losses, while the latter is governed by catalyst layer thickness-scale (or global) losses. The presence of an optimal performance with varying Nafion content in the cathode is more due to the local agglomerate-scale mass transport and conductivity losses in the polymer coating around the agglomerates than due to the amount of Nafion within the agglomerate. It is also found that platinum mass loading needs to be at a moderate level in order to optimize fuel cell performance, even if cost is to be disregarded.

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1. Introduction

One of the impediments to commercial success of fuel cells is high cost. One of the major contributing factors to high cost is the excessive use of platinum within the catalyst layers. Thus, there is strong motivation for using as little platinum as possible without hampering the overall performance of the fuel cell. In a polymer electrolyte fuel cell (PEMFC), since the catalyst (typically platinum) is dispersed within a complex porous matrix comprised of carbon, the ionomer (typically Nafion) and platinum, the overall performance of the fuel cell does not scale linearly with an increase in the amount of platinum within the catalyst layer. Rather, the performance depends on how effectively the platinum is dispersed within the porous matrix so that most of it is actually utilized in catalyzing the electrochemical reactions. Since the proper functioning of a PEMFC cathode requires existence of triple phase boundaries [1,2] between the Nafion (for proton transfer), platinum (for

catalysis) and carbon (for electron transfer), the determination of optimum composition and structure of the catalyst layer is a monumental task. Such studies are often undertaken using experiments [3,4], but are very time-consuming and expensive. Needless to say, with trial-and-error experimental procedures, only a small portion of the overall design space can be explored within a reasonable time. Computational modeling provides an alternative means to address this critical issue. However, such models also require careful formulation and, to some extent, calibration of unknown fitting parameters. Nevertheless, given the importance of the problem in the context of fuel cell technology advancement, and the difficulties associated with experimental trial-and-error procedures, there is strong motivation to try to address the problem using models, either fundamental or phenomenological.

Over the past decade or so, there has been an explosion in the number of technical publications that have reported findings of the simulation of the catalyst layer of a PEMFC. Broadly, these simulations may be classified into two categories: (1) simulations that utilize approximate models based on the pseudo-homogeneous film concept [5–9], or the flooded agglomerate concept [10–24], and (2) direct numerical simulation (DNS) of the catalyst layer [25,26]. While the latter approach is more powerful, it has serious limitations. First, it requires reconstruction of the catalyst layer

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Nomenclature

A_v	total catalyst surface area per unit volume of cathode (m^{-1})
A_0	total catalyst surface area per unit mass of catalyst particle ($\text{m}^2 \text{kg}^{-1}$)
c	dissolved oxygen concentration in Nafion (kmol m^{-3})
c^*	dissolved oxygen concentration in Nafion in equilibrium with inlet gas (kmol m^{-3})
$c_{\text{O}_2, \text{g}}$	oxygen gas concentration in cathode gas pores (kmol m^{-3})
$c_{\text{O}_2, \text{g}}^*$	oxygen gas concentration at cathode inlet (kmol m^{-3})
c_0^{ref}	standard reference oxygen concentration (kmol m^{-3})
C_w	concentration of water in membrane (kmol m^{-3})
\bar{d}	average pore size of cathode (m)
D_{kn}	binary diffusion coefficient of species k into n ($\text{m}^2 \text{s}^{-1}$)
$D_{\text{O}_2, \text{N}}$	diffusion coefficient of oxygen in Nafion ($\text{m}^2 \text{s}^{-1}$)
D_T	temperature dependence of D_λ ($\text{m}^2 \text{s}^{-1}$)
D_w	diffusion coefficient of water ($\text{m}^2 \text{s}^{-1}$)
D_λ	diffusion coefficient of water ($\text{m}^2 \text{s}^{-1}$)
D'_λ	concentration dependence of D_λ
$D_{\text{O}_2, \text{N}}^{\text{eff}}$	effective diffusion coefficient of oxygen in Nafion in agglomerate ($\text{m}^2 \text{s}^{-1}$)
Eff_1	nucleus effectiveness factor
Eff_2	film effectiveness factor
F	Faraday's constant ($96.487 \times 10^6 \text{ C kmol}^{-1}$)
\mathbf{i}	net current density vector (A m^{-2})
\mathbf{i}_F	ionic phase current density vector (A m^{-2})
\mathbf{i}_S	electronic phase current density vector (A m^{-2})
$i_{\text{S}}^{\text{cat}}$	surface current density on a smooth catalyst surface (A m^{-2})
i_0^{ref}	standard exchange current density on cathode (A m^{-3})
j_0	reference current density (A m^{-2})
j_{T}^{an}	net transfer current at anode due to electrochemical reaction (A m^{-2})
$j_{\text{T}}^{\text{cat}}$	net transfer current density at cathode due to electrochemical reaction (A m^{-3})
\mathbf{J}_{adv}	water flux due to advective transport ($\text{kmol m}^{-2} \text{s}^{-1}$)
\mathbf{J}_{diff}	water flux due to diffusion ($\text{kmol m}^{-2} \text{s}^{-1}$)
\mathbf{J}_{drag}	water flux due to electro-osmotic drag ($\text{kmol m}^{-2} \text{s}^{-1}$)
\mathbf{J}_k	diffusion mass flux of the k th species ($\text{kg m}^{-2} \text{s}^{-1}$)
L	cathode catalyst layer thickness (m)
m_{Pt}	platinum mass loading (kg m^{-2})
M_{m}	molecular weight of the membrane (kg kmol^{-1})
M_k	molecular weight of k th species (kg kmol^{-1})
n	number of electrons transferred during the electrochemical reaction
\hat{n}	number of agglomerates per unit volume of cathode (m^{-3})
N	total number of gas-phase species
p	pressure (Pa)
Pt C	platinum–carbon mass ratio in catalyst layer ink
r_{agg}	radius of agglomerate (m)
R	universal gas constant ($8314 \text{ J kmol}^{-1} \text{ K}^{-1}$)
\dot{S}_k	production rate of k th species ($\text{kg m}^{-3} \text{s}^{-1}$)

$(S/V)_{\text{eff}}$	Effective surface to volume ratio (m^{-1})
t	time (s)
T	temperature (K)
\mathbf{U}	Bulk fluid velocity (m s^{-1})
V_{oc}	open circuit voltage (V)
Y_k	mass fraction of k th species

Greek letters

α_a, α_c	Tafel constants for anode
α_T	Tafel constant for cathode catalyst model
β_k	concentration exponents for the k th species
δ	polymer coating thickness around agglomerate nucleus (m)
ε	wet porosity
ε_{agg}	volume fraction of polymer in agglomerate nucleus
ε_{cat}	porosity of cathode catalyst layer
ε_S	volume fraction of platinum + carbon in cathode
ε_N	volume fraction of polymer in cathode
η	electrode overpotential (V)
η_d	electro-osmotic drag coefficient
κ	permeability (m^2)
λ	water content
$[A_k]$	molar concentration of species k (kmol m^{-3})
μ	dynamic viscosity ($\text{kg m}^{-1} \text{s}^{-1}$)
ρ	mass density of mixture (kg m^{-3})
ρ_C	density of carbon (kg m^{-3})
ρ_{Pt}	density of platinum (kg m^{-3})
$\rho_{\text{m}}^{\text{dry}}$	density of dry membrane (kg m^{-3})
σ	electrical conductivity ($\Omega^{-1} \text{m}^{-1}$)
σ_F	electrical conductivity of the ionic phase ($\Omega^{-1} \text{m}^{-1}$)
σ_S	electrical conductivity of the electronic phase ($\Omega^{-1} \text{m}^{-1}$)
σ_{30}	concentration dependence of electrical conductivity ($\Omega^{-1} \text{m}^{-1}$)
τ_{cat}	tortuosity of cathode
Φ_F	ionic phase potential (V)
Φ_S	electronic phase potential (V)

microstructure from SEM or TEM images. Secondly, direct numerical simulations are difficult to perform and are very expensive. Simulation times for a single case may often run into days. Finally, certain compromises have to be made with regard to the actual geometry [25], rendering the results of these studies inconclusive in many cases. Nevertheless, this approach is often useful for fundamental understanding of the coupling between transport phenomena and heterogeneous reactions at the micro-scale and can be used as a framework to develop simplified models.

The more popular approach is based on models of coupled transport and reactions within the catalyst layer structure. Historically, two different model types have been used for this purpose. The first model type, generally referred to in the literature as the pseudo-homogeneous film model [5–9], assumes that the catalyst layer is a porous matrix comprised of Nafion, platinum, and carbon in random (homogeneous) configuration. This model allows for pathways of gases, electrons, and protons within the catalyst layer, and captures some of the essential transport phenomena prevalent in the catalyst layer. However, this model does not acknowledge the necessity for the existence of the triple-phase boundary for a functioning catalyst layer. In contrast, the flooded agglomerate concept, proposed in the late 1980s [10,11], contends that the platinum is supported on carbon particles, which forms agglomerates when mixed with an ionomer. The agglomerate may even be coated fully

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