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Analytical solutions of an isothermal two-dimensional model of a cathode flow channel in a proton exchange membrane fuel cell

Aniket U. Thosar *, Ashish K. Lele

Academy of Scientific and Innovative Research (AcSIR), CSIR-National Chemical Laboratory (CSIR-NCL) campus, Pune, India
CSIR-National Chemical Laboratory, Pune, India

HIGHLIGHTS

- An isothermal 2D analytical model for PEMFC is presented.
- Variation in O_2 concentration and velocity along channel depth is accounted for.
- Model shows excellent match with 3D simulations, as compared with previous work.
- Effect of finite rib width is captured through a correction to the analytical model.

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ABSTRACT

Two key assumptions are usually made while deriving analytical solutions of coupled kinetics and transport equations in a single channel on the cathode plate of a proton exchange membrane fuel cell (PEMFC). These are: plug flow and uniform oxygen concentration along the depth of the channel. However these assumptions are not always valid under typical operating conditions of a PEMFC, and particularly so at high current density. In this article we relax these two assumptions and present approximate analytical solutions of the governing equations using the methodology of separation of variables followed by power series solution. Spatial profiles of oxygen concentration and current density were derived, which led to the final derivation of a comprehensive current-potential relationship (polarization curve) in the reaction-controlled regime of an operational PEMFC. We compare polarization curves predicted by the present model with predictions of the earlier analytical model and also with a complete 3D-simulation of the same flow geometry and operation conditions. The local profiles of oxygen concentration and the polarization curve predicted by the present model compare far better with the 3D simulations than the earlier analytical model. While this comparison highlights the importance of the effects of finite oxygen diffusion rate and velocity profile in the channel on the polarization curves, it also points to other important factors that affect the current-potential relation.

1. Introduction

Proton exchange membrane fuel cell (PEMFC) is evolving into a promising energy delivery device of high efficiency. It directly converts the chemical energy of hydrogen into electrical energy via two half-cell reactions: the hydrogen oxidation reaction (HOR) on the anode side and the oxygen reduction reaction (ORR) on the cathode side. ORR being a sluggish reaction relative to HOR, the anode side processes contribute negligibly in determining the overall power density of a cell (Neyerlin et al., 2005) and therefore extensive modelling and simulation efforts have been invested to understand the physicochemical processes on the cathode side of an operational PEMFC (Cheddie et al., 2005; Djilali, 2007; Weber and Newman, 2004; Weber et al., 2014).

Historically, the complex three-dimensional problem of ORR kinetics coupled with heat, mass and momentum transfer occurring inside a fuel cell has been reformulated into a relatively simpler one-dimensional problem that focuses only on the reaction and transport processes occurring along the thickness of the catalyst layer inside the membrane electrode assembly. This simplification enables better understanding of the factors that govern current density-overpotential relationship (polarization curve) of the fuel cell. It is assumed in these simpler models that the contribution of convection in the flow channel can be neglected, or in

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* Corresponding author at: Academy of Scientific and Innovative Research (AcSIR), CSIR-National Chemical Laboratory (CSIR-NCL) campus, Pune, India.
E-mail address: au.thosar@ncl.res.in (A.U. Thosar).

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Nomenclature

A_c	Tafel slope, (V)
C	local gas concentration in the channel, $(\mathrm{mol/m^3})$
C_{in}	inlet gas concentration, $(\mathrm{mol/m^3})$
$C_{CCL-GDL}$	local oxygen concentration at the CCL-GDL interface, $(\mathrm{mol/m^3})$
C_{Ch-GDL}	local oxygen concentration at the channel-GDL interface $(\mathrm{mol/m^3})$
D	oxygen diffusion coefficient in channel, $(\mathrm{m^2/s})$
D_{CCL}	effective oxygen diffusion coefficient inside the CCL, $(\mathrm{m^2/s})$
D_{GDL}	effective oxygen diffusion coefficient inside the GDL, $(\mathrm{m^2/s})$
F	Faraday's constant, (C/mol)
$2h$	channel height, (m)
$i_{o,c}A$	volumetric exchange current density, $(\mathrm{A/m^3})$
j	local current density, $(\mathrm{A/m^2})$
j_{1-D}	current density as given by one dimensional framework, $(\mathrm{A/m^2})$
j_{flow}	inlet oxygen flow rate driven limiting current density, $(\mathrm{A/m^2})$
j_{lim}	oxygen diffusion inside GDL driven limiting current density, $(\mathrm{A/m^2})$
$j_{diff,ch}$	oxygen diffusion along th depth of the channel driven limiting current density, $(\mathrm{A/m^2})$
L	channel length, (m)
l_{GDL}	thickness of GDL, (m)
l_t	thickness of CCL, (m)
u	local velocity, (m/s)
U_{mean}	mean velocity at the channel inlet
$w_{channel}$	width of channel (m)
w_{rib}	width of ribs (m)
x	co-ordinate along the channel, (m)
y	co-ordinate along the channel depth, (m)

Greek letters

α	dimensionless current density, $\frac{j_{1-D}}{j_{lim}}$
β	dimensionless current density, $\frac{j_{1-D}}{j_{flow}}$
ξ	dimensionless current density, $\frac{j_{1-D}}{j_{diff,ch}}$

other words the oxygen concentration at the interface of the cathode catalyst layer (CCL) and the gas diffusion layer (GDL) is uniform over the entire active area. The first steps in this direction were taken by Bernardi and Verbrugge (1991) and Springer et al. (1993), who presented numerical solutions of a macro-homogeneous model (MH) for the CCL in which protons migrate from the membrane-CCL interface to the CCL-GDL interface while simultaneously reacting with oxygen that diffuses in the opposite direction. Perry et al. (1998) and Eikerling and Kornyshev (1998) elaborated this model further and presented numerical solutions to show how the apparent Tafel slope can double and quadruple because of resistance to oxygen diffusion and proton conduction in the CCL. Kulikovsky (2002, 2010) derived analytical solutions of this model in different regimes of CCL operation namely, ideal ionic transport regime and ideal oxygen transport regime.

The typical boundary condition of uniform oxygen concentration at CCL–GDL interface in one dimensional modelling of CCL may not be true in practical situations. It is intuitively clear that consumption of oxygen in the ORR would reduce its concentration in the oxidant during its travel in the flow channels of the cathode plates. Therefore, oxygen concentration at the CCL-GDL interface should also reduce from inlet to the outlet. This non-uniformity of oxygen concentration gives rise to a current density profile along the channel. Three-dimensional numerical simulations of PEMFC rigorously account for the non-uniform oxygen concentration at the CCL-GDL interface over the entire active area. Some of the earlier work in this area are by Berning et al. (2002, 2003), Dutta et al. (2001), Ju et al. (2005), Mazumder and Cole (2003), and Um and Wang (2004), while a comprehensive review of prior work in this area can be found in Wang et al. (2011). However, analytical treatments of governing equations are sparse even in the limiting cases. The only exception is the work of Kulikovsky (2004) and Kulikovsky et al. (2004) who proposed a two-dimensional isothermal model comprising intrinsic Tafel kinetics in the CCL and oxygen transport resistance across the GDL coupled with material balance equation along flow channel, and provided analytical derivation of the current-potential relationship. However, it was assumed that flow in the channel was plug flow and oxygen concentration was allowed to vary only along the channel length but not along the channel depth. Under typical operating conditions of PEMFC, the flow in channels is laminar and therefore the assumption of plug flow is not realistic. Furthermore as will be seen later, the resistances to oxygen diffusion in the GDL and in the channel are of the same order for a typical fuel cell construction. Hence, the assumption of uniform oxygen concentration along the channel depth is also not realistic. In particular, this assumption can lead to considerable deviations at high current density as will be shown later. A comparison between three-dimensional numerical simulation of governing equations and the two-dimensional analytical model proposed by Kulikovsky et al. (2004) should bring out the criticality of these assumptions and to the best of our knowledge, literature lacks such a comparison.

In this paper the assumptions of laminar flow and uniform oxygen concentration along the channel depth are relaxed and analytical solutions of the revised model are presented. The effects of relaxing the two assumptions are highlighted by comparing the model predictions with those of Kulikovsky et al. (2004) as well as with the results of full three-dimensional numerical simulation of PEMFC under identical operating conditions. Furthermore, the implications of infinitely small rib-width, which is an implicit assumption in the analytical framework, are also discussed thoroughly.

2. Mathematical model

Fig. 1 depicts the two-dimensional domain of the model comprising a channel of length L and depth $2h$ in which laminar flow of the reactant gas occurs along the axial $(x-)$ direction and flow gradient is along the transverse $(y-)$ direction. Channel wall is located at $y = h$ and channel-GDL interface at $y = -h$. No-slip condition is assumed at both boundaries. Because of the reaction in the CCL and the consequent consumption of oxygen, the oxygen concentration in the reactant gas is expected to vary along both x and y directions in the flow channel. Oxygen flux at the channel wall is zero while that at the channel-GDL interface is related to the rate of consumption by ORR in the CCL.

At steady state, transport equation for oxygen in channel can be written as,

$$u\frac{\partial C}{\partial x} = D\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2}\right) \tag{1}$$