Modeling Current Density Distribution Inside Proton-Exchange Membrane Fuel Cells

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A numerical model of the cathode of a proton-exchange membrane fuel cell (PEMFC) based on the discrete geometric approach is presented. The current density distribution inside a PEMFC cathode is simulated. The discrete formulation of a steady-state conduction problem in nonisotropic media is coupled to nonlinear boundary conditions representing the charge transfer at catalyst layers. The cathode performance is assessed under different load conditions by assuming a constant voltage operation mode. Results are in good agreement with those presented in the literature.

Index Terms—**Discrete geometric approach, fuel cell, nonlinear conduction, proton-exchange membrane fuel cell (PEMFC).**

I. INTRODUCTION

DURING the last decade, the idea of a sustainable development based on eco-compatible technologies has drawn increasing attention. Fuel-cell technologies, being based on clean, efficient, and low impact electrochemical processes, have been widely investigated [1], [2]. These devices produce electric power from oxidation of hydrogen or other fuels, which are continuously supplied. Fuel cells are an important technology for a wide variety of applications, including automotive and distributed generation. Proton-exchange membrane fuel cells (PEMFCs) are considered to be the most suitable fuel cells for transportation and portable applications due to their low operating temperature, high energy density, and efficiency.

PEMFCs consist of a membrane electrode assembly (MEA) sandwiched between the anode (ACC) and the cathode current collectors (CCCs) in Fig. 1. The MEA can be divided into five layers compounded together: the anodic/cathodic catalyst layers (ACL and CCL) are dispersed on both sides of the polymeric electrode membrane (PEM), which is interleaved between the gas anodic/cathodic diffusion layers (ADL and CDL). Chemical reactions take place at catalyst layers where three different phases coexist. Triple-phase boundaries (TPBs) are the most active sites for electrode reactions since electrode particles, electrolyte phase, and gas pores intersect. These complex phenomena are quite difficult to be modeled, even though some attempts have been made with approximate models like the one presented in [3].

The reactants flow through diffusion layers from flow channels to the catalyst layers. Oxygen is typically taken from the atmospheric air and hydrogen is stored in high-pressure vessels or in metal hydride cartridges. Electric charges follow different paths: electrons are drawn from the TPB by the anode diffusion layer and flow to the external circuit through current collectors, while protons flow through the PEM. Electrons and protons are consumed in the oxygen reduction reaction producing water.

In this context, mathematical models are useful for interpreting experimental data and designing optimized fuel-cell configurations. A higher current density for a given cell voltage
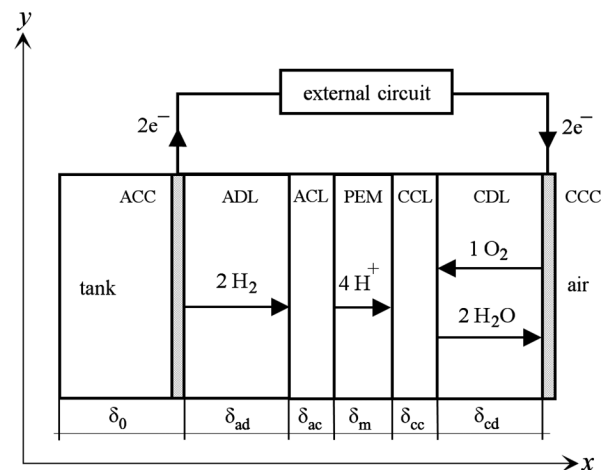

Fig. 1. PEMFC structure: the membrane electrode assembly is sandwiched between the anode and cathode current collectors.

and more uniform current density on the cross section are the current issues under investigation. Several mathematical models have been developed to investigate the effect of various design and operating conditions on the fuel-cell performance and to understand the underlying mechanism, either for solid-polymer-electrolyte or PEMFCs [4]–[6]. Moreover, advanced computational models (multidimensional, multiphase models), which account simultaneously for electrochemical kinetics, current distribution, hydrodynamics, and multicomponent transport may provide a more accurate picture of transport and kinetic phenomena in PEMFCs with different flow distribution strategies [7].

The aim of this paper is to develop a numerical simulation model of the electronic conduction inside the cathodic CDL attached to a segmented graphite current collector. In a typical PEMFC, one of the basic features to be improved is the average current density for a fixed operating cell voltage. On the other hand, uniformity of current density distribution across the entire active area is essential for performance optimization.

The effect of several features, such as electrode configuration, air-flow rate, fuel and oxidant gas stream humidity, and cell temperature on the spatial and temporal current density distribution have been investigated by representing the PEMFC cathode as a segmented electrode/current collector assembly as described in [8] and [9]. The electric scalar potential distribution is simulated by solving a nonlinear, anisotropic conduction problem, where field equations are expressed directly in algebraic form

by means of the discrete geometric approach (DGA) [11], [10]. The current density distribution can be easily computed from potential distribution in order to estimate its degree of uniformity through the entire active area of the electrodes [9]. As a result, it is possible to increase the current density for a given working cell potential.

II. DISCRETE GEOMETRIC MODEL

We propose a 2-D discrete geometric model of the cathode region, where the electronic conduction occurs. Due to the periodic symmetry of the model, only a part of the electrode structure is considered. We denote $D = D_{CDL} \cup D_a \cup D_b$ as the domain of interest, consisting of a CDL region D_{CDL} and one half of two adjacent graphite current collectors plates D_a, D_b separated by an insulating region D_i (Fig. 2).

The upper part of the boundary of the D_{CDL} region is in tight contact with the cathode catalyst layer, where electric charges are consumed due to the oxygen reduction reaction. The cathode catalyst layer is assumed to be a zero-thickness region since its dimension is typically negligible compared to that of the other layers of the MEA.

A. Domain Discretization

According to the DGA, the computational domain D is discretized into a pair of interlocked cell complexes, one dual of the other (Fig. 3). In our 2-D field problem, the current density $\mathbf{J}$ and electric field $\mathbf{E}$ are assumed to be parallel to the xy plane; the primal cell complex consists of nodes n, edges e, faces[1] f, and volumes (prisms with triangular base) v; we denote with N, E, F, and V the cardinalities of the corresponding sets of geometric elements, respectively. The dual cell complex, consisting of dual faces[2] $\tilde{f}$, dual edges $\tilde{e}$, and dual nodes $\tilde{n}$, is obtained according to the barycentric subdivision from the primal complex [11]. Since the problem is 2-D and the current density and electric field lie in the xy plane, we consider the projection on a plane of such a pair of interlocked grids; therefore, the primal volumes v_k correspond to triangles s_k with $k = 1, \ldots, V$, and the primal faces correspond to edges. In the following text, we will denote $\mathbf{G}$ as the incidence matrix of dimension $E \times N$ between primal edges and primal nodes, with $\mathbf{C}$ as the incidence matrix of dimension $F \times E$ between primal faces and primal edges, and with $\tilde{\mathbf{D}} = -\mathbf{G}^T$ representing the incidence matrix of dimension $N \times E$ between dual volumes and dual faces.

B. Nonlinear Discrete Conduction Problem

The behavior of the cathode catalyst layer is simulated by introducing appropriate nonlinear Neumann boundary conditions, which account for the electric charge transfer rate. According to the DGA, the physical laws governing the nonlinear conduction problem inside the cathode region can be expressed *exactly* by means of algebraic relations between degrees of freedom using only the topological information of the mesh embedded in the incidence matrices.

Hence, Faraday's law for stationary fields can be expressed with the DGA directly in algebraic form [12] as

$$\mathbf{CU} = \mathbf{0} \qquad (1)$$

[1]Each surface is of thickness d and its trace is a line in the xy plane.

[2]Each dual surface is of thickness d and its trace is a line in the xy plane.
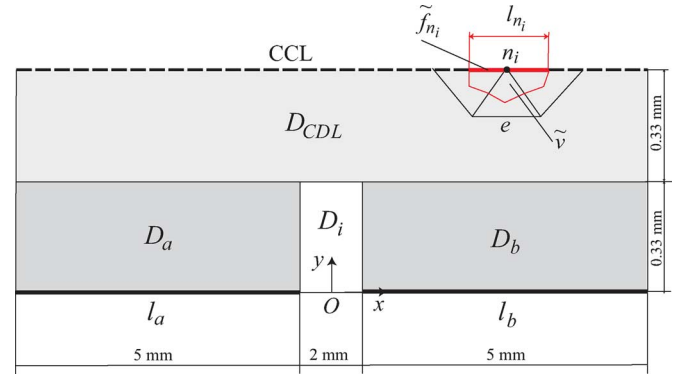

Fig. 2. Two-dimensional model on the (x, y) plane of a segmented electrode at the cathode. (The picture is not scaled proportionally.) The cathode catalyst layer (CCL)—where reactions take place—is evidenced on the top.
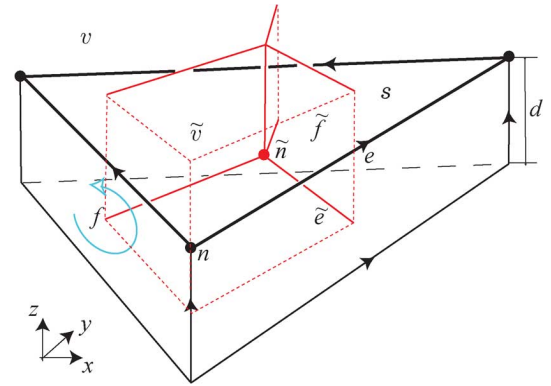

Fig. 3. Geometric entities of primal and dual cell complexes tailored inside the primal volume v in a one-to-one correspondence with triangle s; in particular, primal nodes n and primal edges e are evidenced, since they define a 2-D planar mesh. The thickness of the triangular prism v is denoted by d.

where $\mathbf{U}$ is the array of the voltages $(\mathbf{U})_k = \int_{e_k} \mathbf{E} \cdot d\mathbf{l}$ on primal edge e_k, with $k = 1, \ldots, E$; $(\mathbf{x})_k$ denotes the kth entry of the array $\mathbf{x}$. We define $\mathbf{V}$ as the array of the electric scalar potentials $(\mathbf{V})_i$ associated with each primal node n_i, with $i = 1, \ldots, N$ (Fig. 2). By noting that $\mathbf{CG} = \mathbf{0}$, (1) can be implicitly enforced at the discrete level by means of electric scalar potentials as

$$\mathbf{U} = -\mathbf{GV}. \qquad (2)$$

The current continuity law can be written as the following algebraic relation:

$$-\mathbf{G}^T \mathbf{I} + \mathbf{I}_s(\mathbf{V}) = \mathbf{0} \qquad (3)$$

where $\mathbf{I}$, $\mathbf{I}_s$ are arrays of current of dimension E; $\mathbf{I}_s$ denotes the array of source currents, which depends nonlinearly on the electric scalar potential.

The Ohm's law $\mathbf{J} = \sigma \mathbf{E}$ between fields can be expressed in discrete form with a *constitutive matrix* σ that links the array of voltages $\mathbf{U}$ to the array of currents $\mathbf{I}$ as

$$\mathbf{I} = \sigma \mathbf{U}. \qquad (4)$$

The square matrix σ of dimension E encodes the information of the material properties and of the geometry of the problem. The constitutive matrix σ can be easily constructed in a geometric fashion (i.e., by applying for triangles the same ideas developed

in [12] and [13] for tetrahedra) in such a way that (4) holds exactly for an *element-wise uniform* electric field E, electric current density J, and conductivity tensor σ_k in each volume v_k.

C. Modeling the Catalyst Layer

The current density $J(n_i)$ in a neighborhood of each node n_i in the catalyst layer can be modeled according to the Butler–Volmer equation accounting for, for example, the PEM hydration, the reactant and product concentration, and temperature [8], [9]

$$J(n_i) = -J_0 \left[10^{\frac{(\mathbf{V})_i - E^0}{\eta_c}} - 10^{-\frac{(\mathbf{V})_i - E^0}{\eta_c}} \right] \quad (5)$$

where E^0 is the equilibrium potential characteristic of the electrochemical reaction, η_c is the cathode Tafel constant, and J_0 is the apparent exchange current density. By multiplying the current density $J(n_i)$ by the area $|\tilde{f}_{n_i}| = dl_{n_i}$ of the dual face $\tilde{f}_{n_i}$, the corresponding source current $(\mathbf{I}_s)_i$ is obtained, where d is model thickness and l_{n_i} is the $\tilde{f}_{n_i}$ width (Fig. 2). Thus, $(\mathbf{I}_s((\mathbf{V})_i))_i$ is a prescribed function of the electric potential $(\mathbf{V})_i$ in the node n_i. If the node n_i belongs to the catalyst layer, then $(\mathbf{I}_s(\mathbf{V}))_i$ is the current crossing the dual face $\tilde{f}_{n_i}$ corresponding to the node n_i; the entries of $\mathbf{I}_s(\mathbf{V})$ are null for any other node not belonging to the catalyst layer.

III. NONLINEAR COUPLED PROBLEM

By inserting (2) and (4) in (3), the following algebraic system of equations is obtained:

$$(\mathbf{G}^T \boldsymbol{\sigma} \mathbf{G} \mathbf{V})_i + (\mathbf{I}_s(\mathbf{V}))_i = 0, \quad i = 1, \dots, N. \quad (6)$$

The global stiffness matrix $\mathbf{G}^T \boldsymbol{\sigma} \mathbf{G}$ is obtained by assembling the contributions from the local stiffness matrices of each triangle s_k in a one-to-one correspondence with a primal volume v_k, with $k = 1, \dots, V$. The entry $(\mathbf{G}^T \boldsymbol{\sigma} \mathbf{G})_{lm}^k$ of a local symmetric stiffness matrix can be expressed efficiently [12] in a pure geometric way for the triangle s_k as

$$(\mathbf{G}^T \boldsymbol{\sigma} \mathbf{G})_{lm}^k = \frac{1}{4|s_k|} \mathbf{f}_l \cdot \sigma_k \mathbf{f}_m, \quad l, m = 1, \dots, 3 \quad (7)$$

where $|s_k|$ is the area of s_k, and $\mathbf{f}_l$ is the area vector[3] associated with the lateral face f_l of v_k. In the CDL region D_{CDL}, the local conductivity tensor σ_k in v_k is diagonal, but it represents an anisotropic conductivity along orthogonal directions, while the conductivity of the bulk graphite collector regions (D_a, D_b) is isotropic.

Dirichlet or Neumann linear BCs must be considered in addition along with l_a, l_b lines to ensure the uniqueness of the solution (Fig. 2). For constant voltage operation, the potential is imposed to a fixed value; for constant current operation, a given uniform current density along l_a, l_b is imposed and, consequently, the potential V_{l_a}, V_{l_b} of the equipotential interfaces l_a, l_b is computed; in both cases, the potential is unknown along the catalyst layer. Finally, symmetry BCs on the lateral sides of region D are applied.

IV. NUMERICAL RESULTS

The potential distribution in the cathodic region of a segmented electrode cathode of a PEM fuel cell is analyzed by considering a portion of the periodic current collector structure de-

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[3]It is normal to the lateral face f_l of the prism and it points outward v_k; moreover, due to the plane symmetry, $|\mathbf{f}_l| = |\mathbf{e}_l|$ holds.

In–plane conductivity σ_{xGDL}	78.5 S/cm
Through plane conductivity σ_{yGDL}	3.14 S/cm
Bulk conductivity of graphite collectors $\sigma_{Da,b}$	670 S/cm
Equilibrium potential E_0	1.2 V
Cathode Tafel constant η_C at 30°C	69.3 mV/dec
Exchange current density J_0 at 30°C	5 nA/cm^2
η_a at 30°C	40mV cm^2/A

picted in Fig. 2. The geometric data and the electric parameter values reported in Table I refer to the fuel cell described in [9].

We solve the nonlinear system (6) by means of a Newton–Raphson method. The Jacobian can be evaluated from the residual of the nonlinear system

$$\mathbf{f}(\mathbf{V}) = \mathbf{G}^T \boldsymbol{\sigma} \mathbf{G} \mathbf{V} + \mathbf{I}_s(\mathbf{V}) \quad (8)$$

as

$$(\mathbf{J})_{ij} = \frac{\partial (\mathbf{f}(\mathbf{V}))_i}{\partial (\mathbf{V})_j} = (\mathbf{G}^T \boldsymbol{\sigma} \mathbf{G})_{ij} + \frac{\partial (\mathbf{I}_s((\mathbf{V})_i))_i}{\partial (\mathbf{V})_j} \delta_{ij} \quad (9)$$

where $(\mathbf{x})_{ij}$ denotes the element of the matrix $\mathbf{x}$ in the ith row and jth column, and δ_{ij} is the Kronecker delta function.

The flowchart of the iterative solution scheme adopted for the solution of the nonlinear system (6) is reported in Fig. 4. In order to improve the convergence rate, we also experimented with a locally linearized expression of the electron current $(\mathbf{I}_s)_i$ across a dual face $\tilde{f}_{n_i}$ instead of (5)

$$(\mathbf{I}_s((\mathbf{V})_i))_i = -\left| \tilde{f}_{n_i} \right| \frac{((\mathbf{V})_i - E^0)}{\eta_a} \quad (10)$$

where $\eta_a = 40$ V cm^2/A.

As a boundary condition, we implemented the so-called constant voltage operation of the fuel cell, where the potential of the primal nodes on l_a, l_b is imposed on $V_{l_a} = V_{l_b} = 0.695$ V, which is the normal voltage operation of the cell described in [9]. The domain D is discretized with a very refined mesh (85 347 nodes, 168 094 triangles). The overall simulation time is about 10 s, including the preprocessing, the $\mathbf{V}^0$ computation, two Newton–Raphson steps and postprocessing. Figs. 5 and 6 show the resulting overall potential and current density distribution at the cathode for a constant voltage operation mode of the fuel cell.

These results are in the same order of magnitude with those reported in the literature obtained by means of a finite difference method [9]. Since contact resistances, which deeply affect numerical and experimental results, are not implemented in our model, a more direct comparison cannot be carried out. Results concerning the constant current operation are not provided in this paper for the sake of brevity. In that case, instead of fixing the potential of the primal nodes on l_a and l_b by means of Dirichlet BCs, the total current crossing dual faces—in one-to-one correspondence with primal nodes on l_a and l_b—is known and can be imposed by means of Neumann BCs.

V. CONCLUSION

The developed coupled model is suitable for analyzing electronic conduction in the cathode current collectors of PEMFCs. The DGA makes use of integral variables, which makes it possible to implement in a straightforward manner nonlinear Neumann BCs. In fact, primal nodes—where potential is associ-
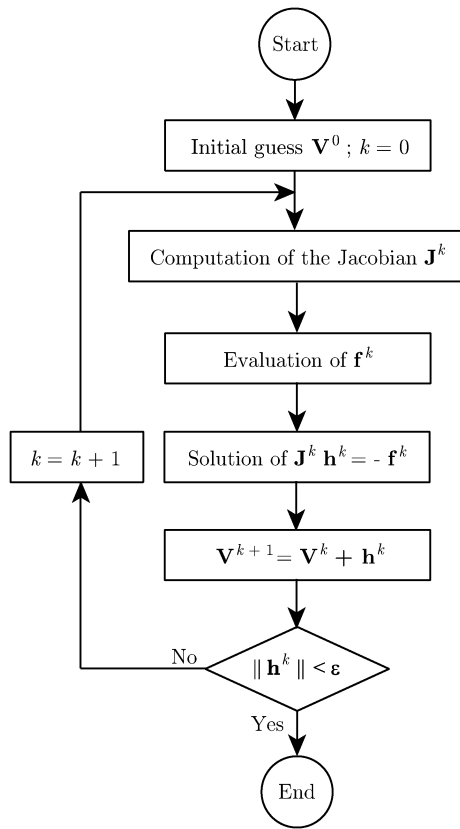
Fig. 4. Flowchart of the iterative Newton–Raphson scheme used for solving the nonlinear system.
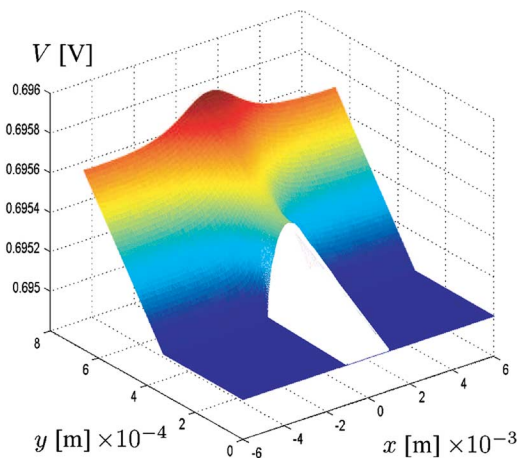

Fig. 5. Potential distribution calculated with the DGA for a constant voltage operation of the fuel cell. The top part of the plot shows the potential distribution at the catalyst layer.
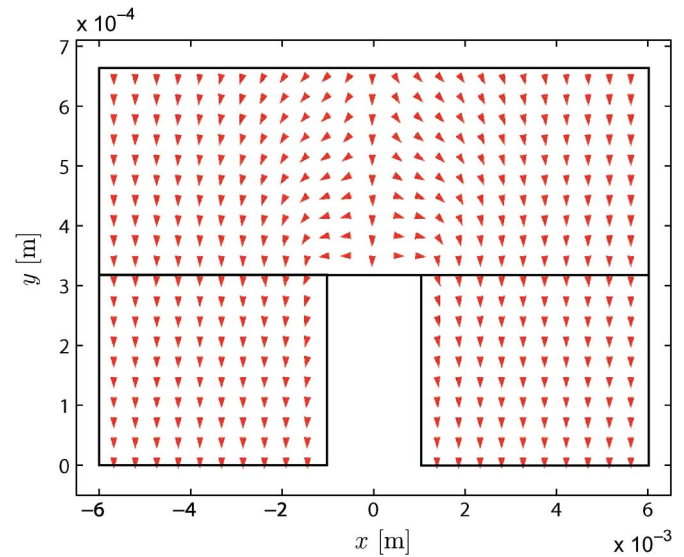

Fig. 6. Planar distribution on the (x, y) plane of the electronic current density distribution in the cathode of the fuel cell computed at constant voltage operation.

ated—and dual faces—where currents are associated—are in one-to-one correspondence as can be observed in Fig. 2. This peculiarity makes the coupling natural between the classical current conduction model and the Butler–Volmer equation. The results obtained with the DGA are in the same order of magnitude with those presented in [9], obtained by means of a finite difference method, and show overall good agreement in terms of qualitative behavior. A further development will be the inclusion of contact resistances in the discrete model.

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