NEW PEM FUEL CELL STACK MODEL CONSIDERING REACTANTS DIFFUSION EFFECTS

Suk Heung Song¹, Song-Yul Choe², Yuyao Shan², Jong Hoon Jang³

¹: Korea Institute of Energy Research, Daejeon, Rep. Korea
²: Department of Mechanical Engineering, Auburn University, Auburn, AL 36849
³: School of Mechanical and Automotive Engineering, University of Ulsan, Ulsan, Rep. of Korea

ABSTRACT

The focus of this paper is placed on research and development of a control-oriented dynamic PEM (polymer electrolyte membrane) fuel cell system model that includes a stack model and system models. Particularly, the stack model considers a single cell model composed of different layers. The framework models for the system are based upon inertia dynamics of compressor, manifold filling dynamics and a simplified humidifier. The stack model is integrated into the system model under MATLAB/SIMULINK environment. Comparative simulation studies demonstrate different dynamics at a varying load than commonly employed semi-empirical models, which are mainly caused by the dynamics of the reactants in the GDL (Gas Diffusion Layer). One of possible applications will be for designing a fuel cell controller.

INTRODUCTION

PEM fuel cell is a potential candidate that can replace current internal combustion engine. Modeling and simulation of fuel cell provide scientific means and methodologies to fundamentally understand the physical mechanisms and to optimally design a fuel cell powered propulsion system. Current available models can be classified in two categories, for designing the layers of a single cell with a high resolution analysis capability and for optimizing system components and controls in conjunction with BOP (Balance of Plant) and power system. The former generally based upon computational fluid dynamics requires intensive computation time and more than 30 physical parameters to be measured. In addition, a deep understanding and wide knowledge in the electrochemical mechanisms are necessary, which impedes practical utilization of the models for a system analysis. The latter, however, is based upon the static behavior of a fuel cell that represents the typical I-V polarization curve. The parameters of equations are determined by fitting empirical values. One of the drawbacks is the lack of dynamics crucial for designing controls and optimizing the system components. Most authors in recent publications are using empirical models that focus on the fuel cell performance with respect to a selection of cell components and an optimization of operating conditions[1][2][3].

Most realistic dynamic models for the PEM fuel cell system have been proposed to design advanced controls and optimize system efficiency[4]. The model considers the
flow characteristics in the fuel delivery that include inertia dynamics of compressor and the manifold filling dynamics. However, the stack model is based on a semi-empirical equation fitting to a polarization curve. Consequently, the dynamic characteristics of the cell have not considered in the model. Therefore, a new model for the PEM fuel cell system is developed to take the dynamics of a cell into account, which is based on electrochemical knowledge in reactions and fluid dynamics of reactants at anode/cathode GDL, catalyst layers and membrane[5].

**PEM FUEL CELL SYSTEM MODEL**

The PEM fuel cell system consists of four subsystems, hydrogen and air supply system, cooling and humidification system, which schematic diagram is shown in Fig. 1.

![Fig. 1 Configuration of PEM Fuel Cell System](image)

Following assumptions are made to simplify the modeling; 1) Cooling and humidification system are perfectly controlled, so that the operating temperature in the stack is constant and the membrane is fully saturated. The corresponding relative humidity of the gas exceeds 100% and vapor condenses into liquid form; 2) All gases obey the ideal gas law and the flow in and out of the manifold is laminar, so that manifolds are described with lumped parameters; 3) The flooding at the diffusion and catalyst layers is neglected.

**Models for System Components**

The model for the system components contains three sub-models for a compressor, a manifold and a stack, whose block diagram is shown in Fig. 2.

1) **Compressor and Manifold.** The compressor model considers the compressor and motor. The air flow rate is determined by a static compressor map, while the exhaust air temperature and compressor power are calculated by a thermodynamic equation. The manifold model describes a lumped volume associated with pipes and connections between the stack and system components. As matter of fact, the inlet temperature in the manifold is different from that of outlet manifold. The inlet temperature rises when the pressure after the compressor gets high, while the changes of air temperature in the outlet
manifold are negligible because of relatively small difference in the pressure drop between the outlet manifold and atmosphere [4].

Fig. 2 Models for System Components

2) Stack. The stack is based on a single cell model multiplied with numbers of cells. A schematic diagram of a cell is depicted in Fig. 3. The models for each layer are governed by the equations given in Table 1, under assumption that the plane of layers is homogeneous.

Fig. 3 Scheme of a Single Cell
Table 1. Governing Equations for Layers in a Cell

| Layer         | Governing Equation          | Analyze Object                                      |
|---------------|-----------------------------|------------------------------------------------------|
| Gas Diffusion | Continuity EQ               | multi-phase/component onvection-diffusion flow in porous media |
|               | Stefan-Maxwell Conservation EQ |                                                       |
| Catalyst      | Reaction-diffusion          | diffusion-reaction flow                               |
| Membrane      | Nernst-Planck               | proton and water transfer                            |

The over-potentials are resulted from the anode and cathode interfacial kinetics, the proton conductivity in the membrane, the cathode mass transport limited by the flooding. As a result, the cell voltage can be expressed as a function of different physical variables like current, anode and cathode pressure, reactant partial pressures, temperature, and membrane resistance. The equations are as follows;

**Electrochemical reaction**

$$V_{cell} = E_o - V_{ohm} - \eta$$

$$V_{ohm} = j_r \cdot R_{mem}$$

where, $E_o$: the open circuit voltage (OCV) has been defined by Nernst equation; $\eta$: the catalyst layer overpotential which can be calculated by using Butler-Volmer equation

$$j_r = j_o A_e \left\{ \frac{p_{H^+}}{p_{H^+}} \right\} \exp \left( \frac{\eta}{b} \right) - 1$$

**Gas diffusion**

The air mainly contains nitrogen, oxygen and water vapor. The oxygen diffuses through the diffusion layer and reaches to the catalyst layer. This phenomenon is described by the mass conservation and Stefan-Maxwell diffusion equations.

$$\frac{\varepsilon_s}{RT} \frac{\partial p_i}{\partial t} + \frac{\partial N_i}{\partial y} = 0; \text{Mass conservation}$$

$$\frac{\varepsilon_s}{\tau^2} \frac{\partial p_i}{\partial y} = \sum_{k=1}^{3} \frac{RT}{p_k \varepsilon_{ik}} (p_i N_k - p_k N_i); \text{Stefan-Maxwell}$$

where, $i, k \in \{1, 3\}; p_1, p_2, p_3$: oxygen partial pressure, vapor partial pressure and nitrogen partial pressure, respectively; $p_k \varepsilon_{ik} = D_{ik}$: diffusivities; $\varepsilon_s$: the diffusion layer porosity; $\tau$: the tortuosity. According to equations above and consideration of only oxygen partial pressure, a following equation can be obtained;

$$\frac{\partial p_1}{\partial t} = \sigma \frac{\partial^2 p_1}{\partial \xi^2} - \psi N_{1dc} \frac{\partial p_1}{\partial \xi}$$

where, $\xi = \frac{y}{L_d}$: dimensionless diffusion layer length; $N_{1dc} (= \frac{j_r}{4F})$: oxygen flow at the interface diffusion-catalyst layers.
Catalyst layer

The proton concentration in the catalyst layer is dependent on the cell current. When the cell current rises, the water production and the hydration of the membrane near the catalyst tend to rise as well, which lead to an increase in the number of mobile protons. Consequently, a high proton concentration near the cathode catalyst layer will be occurring. In contrast, the proton concentration at a rapid decrease of the current does not follow directly the current because of the slow dynamics caused by the excess water that should be removed in the cathode catalyst layer. The behavior of proton concentration is defined by the following equation;

$$ u \left( - \frac{\partial c_{H^+}}{\partial t} \right) \frac{\partial c_{H^+}}{\partial t} + \frac{c_{H^+} - \alpha_{H^+} \cdot j^3 - 1}{\tau_{H^+}} = 0 $$

where,

$ c_{H^+} \left( = \frac{[H^+]}{[H^+]} \right) $: dimensionless proton concentration

$ \tau_{H^+} $: the time constant related to the $ c_{H^+} $ dynamics

$ \alpha_{H^+} $: parameter that links $ c_{H^+} $ to the cell current

SIMULATION AND RESULTS

The behaviors of gases in the fuel supply system are described with mass and energy balance and stoichiometric relationship between reactants. Specially, the fluid flow in the manifold is governed by the filling dynamics. The operation conditions include cell temperature, air pressure in inlet of cathode, oxygen partial pressure, oxygen concentration and membrane humidity. The current serves as a load. The necessary data for model are obtained from different references [4] [5].

For the simulation, following conditions have been assumed: 1) hydrogen supply is relatively fast; 2) hydrogen flow rate can be instantaneously adjusted to provide minimum pressure difference across the membrane; 3) Stack pressure is equal to the anode and the cathode pressure; 4) Operating temperature is at 80°C; 5) Excess oxygen ratio is ranged from 2 to 3; 6) Step changes of load current are from 100A to 180A with several steps. Fig. 4 shows I-V polarization curves dependent on pressure. A comparison between the proposed model and a semi-empirical model illustrates different I-V curve by an increased current density. Particularly, the influence of the flooding effects can be recognized at the high currents.

Fig. 5 and 6 demonstrate different dynamic behavior of fuel cell system with the proposed and the semi-empirical stack model. The output voltage of the stack drops rapidly when the output current increases. After the current had increased, the oxygen excess ratio drops because of a depletion of oxygen. The overall difference in the polarization curve of two models significantly impacts the voltage responses. The dynamic effects of different proton concentrations at the cathode catalysts indicate the state of excess water removal at the catalysts layer. Those are the main factors influencing the different responses.

Fig. 7 shows the variation of oxygen partial pressure in GDL depending on the thickness ($ \zeta = L_d/y $). The output voltage is varying with the parameter, while the semi-empirical model shows no variation of the voltage dependence on the GDL geometry.
Fig. 4: Polarization Curve

Fig. 5: Simulated waveforms of current and voltage
Fig. 6: Simulated waveforms of pressure and oxygen excess ratio

Figure 7: Oxygen partial pressure variation according to GDL thickness
CONCLUSIONS

The proposed model for the PEM fuel cell system includes a cell based on the layers model and system sub-component models. The dynamic characteristics are realized by considering proton concentration in the catalysts layer and oxygen partial pressure in GDL. A comparison between a semi-empirical model and the proposed shows an immense difference in the dynamic responses. The thickness effects of the new models on the response are also simulated. This model will proved a new tool for not only optimizing system components but also designing a fuel cell and a power control in an advanced propulsion system to substantially increase reliability and fuel efficiency of the future power source. However, more analysis will be conducted to improve the dynamic models.

ACKNOWLEDGEMENTS

This work was supported by the Korea Science and Engineering Foundation (KOSEF).

REFERENCES

1. T.E. Springer, T.A. Zawodzinski and S. Gottesfeld (1991). Polymer Electrolyte Fuel Cell Model, J. of Electrochemical Society, v. 138, pp. 2334-2342.
2. D.M. Bernardi and M.W. Verbrugge (1992). A Mathematical model of the solid polymer electrolyte fuel cell, J. of the Electrochemical Society, v. 139, pp. 2477-2491.
3. J.H. Lee and T.R. Lalk (1998). Modeling fuel cell stack systems, J. of Power Sources, v. 73, pp. 229-241.
4. J.T. Pukrushpan, A.G. Stefanopoulou, H. Peng (2002). Modeling and Control for PEM Fuel Cell Stack System, Proceedings of the American Control Conference, Anchorage, AK, pp. 3117-3122.
5. M. Ceraolo, C. Miulli and A. Pozio (2003), Modeling static and dynamic behaviour of proton exchange membrane fuel cells on the basis of electro-chemical description, J. of Power Sources, v. 113, pp. 131-144.