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Title: THROUGH-THE-ELECTRODE MODEL OF A PROTON EXCHANGE MEMBRANE FUEL CELL WITH INDEPENDENTLY MEASURED PARAMETERS

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**THROUGH-THE-ELECTRODE MODEL OF A PROTON EXCHANGE
MEMBRANE FUEL CELL WITH INDEPENDENTLY MEASURED
PARAMETERS**

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A one dimensional model for a proton exchange membrane fuel cell was developed which makes use of independently measured parameters for predicting single cell performance. Optimization of catalyst layer formulation and properties and are explored. Impact of temperature and cathode pressure upon system performance was investigated.

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Introduction

Development of a comprehensive mechanistic model is essential for understanding single cell performance and the variables which impact operation. Both through-the-electrode and down-the channel models have been developed to leverage our experimental effort toward improving membrane electrode assemblies (MEAs) and defining optimum operating conditions. In this presentation, model formulation will be discussed followed by model validation and application to MEA optimization. The impact of cell temperature and operating pressure will also be explored.

Model Formulation

Components of the model consist of the cathode catalytic layer, representation of water transport through the MEA, and oxygen transport through the cathode backing layer. The cathode catalyst layer was initially modeled after Springer et. al.¹ The catalyst layer was represented as a uniform structure with carbon black particles in contact with a continuous ionomer film covering these particles. Void volume within the structure allowed access of oxygen. Equations include the Butler-Volmer equation to describe oxygen reduction kinetics, potential loss through ionic conduction, and oxygen diffusion from the cathode backing layer.

Water transport is modeled through the whole MEA structure. Diffusive transport of gases including water vapor is represented by the Stefan-Maxwell equation in the anode and cathode backing. It is formulated to account for the presence of liquid water in the cathode backing near the catalytic layer. The plane in the cathode backing where water is completely vaporized under some conditions is

calculated. Water transport through the membrane is represented by electro-osmotic drag and diffusive transport.

Input values for parameters are determined from independent measurements as much as possible. Fractional volumes of the various constituents in the catalytic layer are determined by formulation and measurement of the final layer thickness. Oxygen reduction kinetics^{2,3}, water diffusion and electro-osmotic drag⁴, and other parameters were determined from the literature. Only the water saturation within the catalyst layer and cathode backing were input as adjustable parameters with bounds determined from the literature for porous media. Solution of both the catalytic layer current distribution and water transport through the MEA was obtained by integrating with a Runge-Kutta routine with adaptive stepsize control.⁵

Experimental

A single cell and reference test stand were designed and constructed to provide tight control on operating parameters. Key components included mass flow control of gas streams, humidification with HPLC pumps and vaporizers, and balances to weight recovered anode and cathode water. Cell temperature was controlled by water circulation through heat exchangers within the single cell. A data acquisition and control system allowed unattended operation with programmed gas stoichiometries and humidification.^{6,7}

Results and Discussion

Single cell performance was measured as a function of temperature, pressure, and anode and cathode humidification levels. Fig. 1 compares measurements with model results where only water saturation was varied to obtain the match. After tuning the model with this variable, the influence of operating conditions as well as catalyst layer composition and properties could be explored to provide guidance toward optimal configurations.

Two key parameters in MEA optimization include carbon black packing density and NAFION® to carbon ratio within the catalyst layer. Carbon packing density is determined through images of MEA cross sections after testing is complete. In Fig. 2, the relationship between

cell voltage and cathode catalyst loading is explored at three catalyst layer thicknesses as a function of packing density. An optimum is seen in packing density at each layer thickness. The impact of NAFION® loading was also predicted and will be discussed.

Operating temperature and pressure affects overall system performance. As shown in Fig 3, a slight improvement in cell voltage is predicted at elevated temperature. Cell voltage is more strongly dependent on cathode pressure than cell temperature. Air compression energy penalties, however, need to be subtracted to optimize system performance. Fig. 4 illustrates the predicted net cell voltage versus cathode pressure and air stoichiometry. Stoichiometry variations are represented by changes in average cathode gas composition.¹ Improved system performance will be obtained with this MEA formulation at low pressures and moderate stoichiometries.

Conclusion

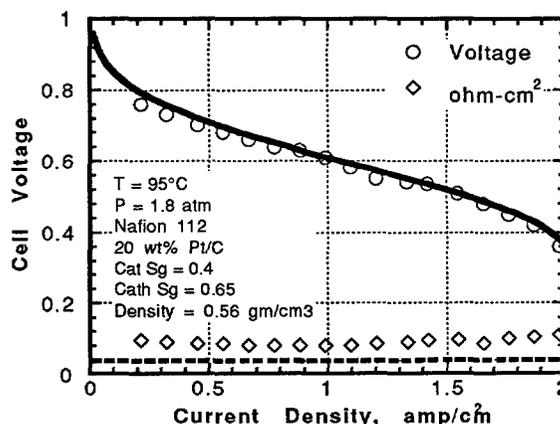
A through-the-electrode model has proved valuable in understanding the key parameters which limit MEA performance. A mechanistic model provides guidance toward optimizing MEA structure and operation conditions.

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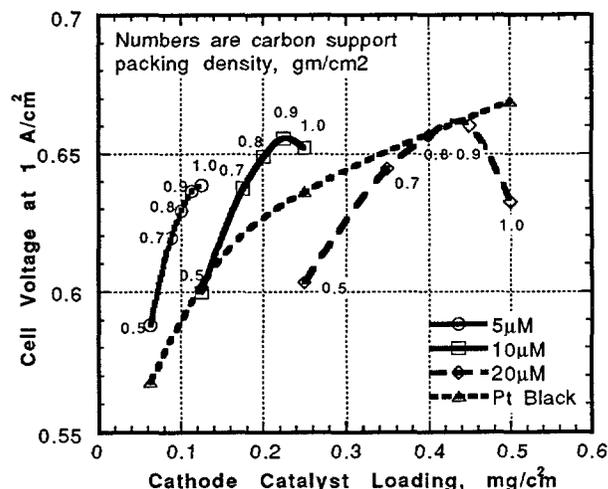
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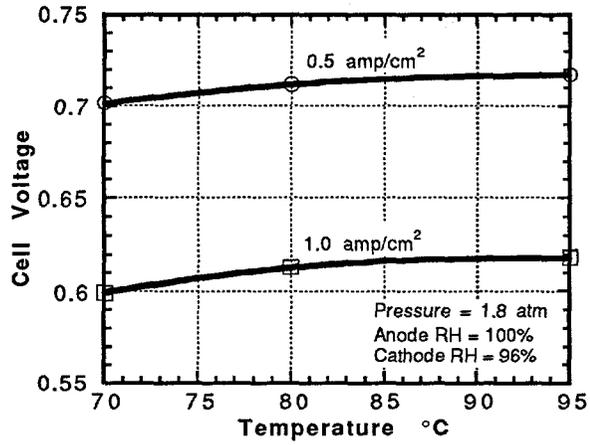
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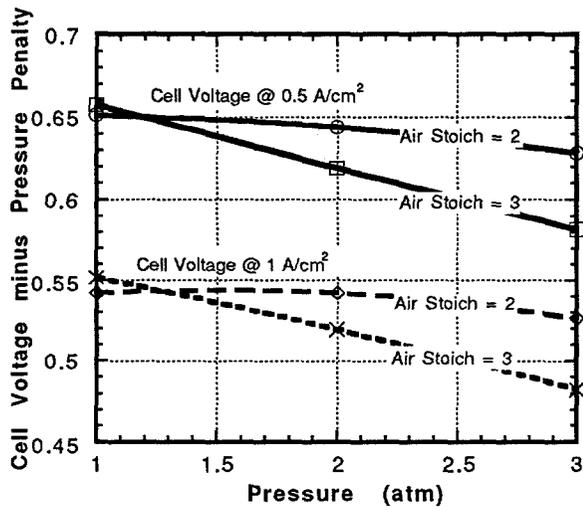
1. Validation of Model with experimental results, 80°C, 1.8 atm, ARH=100%, CRH=96%



2. Predicted Dependence of Cell Performance upon Catalyst Layer Thickness and Density for 20 wt% Pt/C and Platinum Black Catalysts, 95°C, 1.8 atm.



3. Predicted Impact of Cell Operating Temperature Upon Cell Voltage



4. Predicted Impact of Operating Pressure and Cathode Stoichiometry Upon Cell Voltage Corrected for Compressor Pressure Penalty, 80°C, 70% Adiabatic Compressor Efficiency