Two-Layer Model of Diffusional Resistance in the Non-Reacting Gas Diffusion Layer of Elevated Temperature PEMFCs

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The effect of the gas diffusion layer (GDL) on performance of a PEMFC at high current density is assessed using a one-dimensional, multi-component model of diffusion in a non-reacting, porous gas diffusing media on the cathode side. The GDL is presumed to have two distinct regions, as shown in Fig. 1: the high porosity carbon paper substrate, and the lower porosity region of the microporous layer. Typically, the microporous layer is composed of fine carbon particles and a hydrophobic bonding agent. Values of porosity and tortuosity for each of the two discreet layers of the GDL are found by a characterization method that is independent of the electrochemical data.

An exact solution for concentration of oxygen as function of position is found using the Stefan-Maxwell formulation for diffusion. The experimental conditions examined here are for 5 cm^2 cells at 80 ^oC cell temperature with 75% cathode inlet relative humidity, and elevated cell temperatures of 100–120 ^oC. Liquid water is assumed to be absent from the porous material. The diffusion flux used in the model is calculated from limiting current, which is found after applying the appropriate corrections[1] to the polarization data. The limiting current of a cathode is found by extrapolation of current density to zero voltage on the polarization curve. The oxygen concentration at the cathode catalyst goes to zero at the limiting current.

Characterization of the GDL is accomplished by mercury porosimetry, to measure the pore volume as a function of pore diameter, and by gas permeability measurements, stated in the form of the Gurley number. To understand the contribution of each layer to the diffusional resistance, distinct values of porosity and tortuosity are found for each layer. To accomplish this porosimetry measurements of both the composite GDL and the bare carbon paper substrate used in the manufacture of the GDL are required. From consideration of the pore volume available in each of the two discreet regions of the composite gas diffusion layer, a method is derived to construct the pore size distribution of the microporous layer alone using the porosimetry data available from both the bare carbon paper and the composite GDL. The effective depth of penetration of the microporous layer material into the bare carbon paper is determined using the two pore volume measurements.

Porosity is found from the pore volume measurement and the sample geometry. An expression for tortuosity is derived from a model of viscous flow in a capillary tube bundle, the Gurley number and an integration of a function of the pore size distribution. Therefore, the pore size distribution is an essential piece of information in determining the tortuosity. Both in-house and commercially available GDLs are characterized.

Results of the porosity measurement and tortuosity calculation show the porosity of the carbon paper substrates is always relatively high (0.85-0.75), the porosity of the microporous layer is typically lower (0.45-0.30), and the tortuosity of the microporous region is always greater than the bare carbon paper. Qualitatively, the results are not surprising. What is significant is that the single value of porosity, i.e. the ratio of pore volume to total volume of the sample, is not relevant to the calculation of tortuosity. The most significant factor affecting the tortuosity is the pore size distribution. Other investigators[2] have reported the importance of the pore size distribution.

Using the model to calculate oxygen mole fraction as a function of position through the two layers of the GDL, with the values of porosity and tortuosity found by the above technique, reveals the effect of the parameters of each layer on the overall diffusional resistance. For the GDLs evaluated here the resistance to diffusion in the microporous layer is significantly greater than the bare carbon paper and the thickness of each layer is important. By computing limiting current from experimental data for numerous MEAs, with GDLs that have been characterized as described above, the significance of diffusional resistance in the GDL to the oxygen mass transport limitation can be better understood.

If, at limiting current, the only resistance to mass transport is due to molecular diffusion, then the diffusion model presented here gives a reasonable prediction of the oxygen mole fraction as a function of position in the gas diffusion layer. Evaluation of experimental data from this study, along with some preliminary work by the authors, suggests other mechanisms of oxygen transport at high current densities may be significant for certain combinations of GDL material in the MEA, such as convection in the GDL, Knudsen diffusion, and diffusion in the ionomer film at the cathode catalyst surface.

REFERENCES

1. M.V. Williams, E. Begg, L.J. Bonville, H.R. Kunz, J.M. Fenton, Abstracts of the 202nd Meeting of the Electrochemical Society, Symposium S1, Salt Lake City, October 2002.
2. C. S. Kong, D. Y. Kim, H. K. Lee, Y. G. Shul, T. H. Lee, *J. Power Sources*, 4725, 1, (2002)

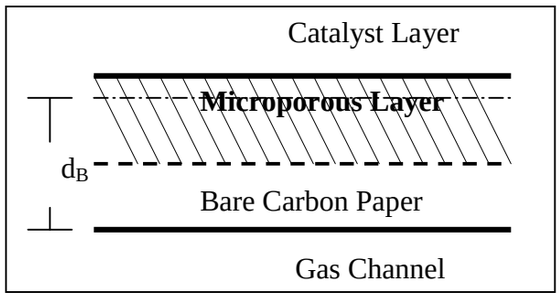

Fig. 1. Schematic of the Gas Diffusion Layer as two regions of distinct porous properties, where d_B is the original thickness of the carbon paper substrate material.