Oxygen Mass Transfer Coefficient Separation into Gas and Electrolyte Contributions in PEMFC GDEs

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The performance of proton exchange membrane fuel cells (PEMFCs) is limited by several processes: the electrochemical kinetics of oxygen reduction, ohmic losses, and the transport of reactants and products within gas diffusion electrodes (GDEs). Transport processes are a major concern for operation at low voltage or high current and for the cathode, because air only contains 21% of oxygen. Oxygen transport from the flow field channel to the catalyst surface occurs by diffusion in gas as well as electrolyte phases. The understanding of transport processes within the GDE is critical for successful improvement and adaptability of PEMFCs to practical operating conditions. In our previous work, we presented a method for the determination of the oxygen mass transfer coefficient by measuring the limiting current density distribution using a segmented cell system. The validity of the mathematical model was also demonstrated (1, 2). The subsequent method development steps include the separation of the overall oxygen mass transfer coefficient into gas and electrolyte contributions and its application to the membrane/electrode assembly (MEA) characterization.

A HNEI's segmented fuel cell and data acquisition system was used (3). A commercially available 100 cm^2 MEA was also used with anode and cathode Pt loadings of 0.1 and 0.2 mg$_{Pt}$ cm^{-2} respectively. A segmented 25BC SGL gas diffusion layer (GDL, 10 segments of 7.6 cm^2) and a Teflon gasket were used at the cathode whereas a single GDL piece and a Teflon gasket were used at the anode. The cell was operated at 60°C, 48.3 kPa$_g$ back pressure and 100% relative humidity. We used He, Ne, N$_2$, Ar, CO$_2$, CF$_4$, SF$_6$ and C$_3$F$_8$ as inert oxygen diluents.

For the case of a dilute reactant stream, the use of a local mass balance leads to the following limiting current density i_l (A m^{-2}) distribution expression (Eq. 1):

$$i_l(y) = \frac{nFkp_r f_r}{RT} e^{\frac{-nFkp_r y}{RTi_e f}} \qquad [1]$$

where y is the dimensionless position along the flow field length, n is the number of electrons exchanged in the electrochemical reaction, F is the Faraday constant (96500 C mol^{-1}), k is the overall mass transfer coefficient (m s^{-1}), p_r is the dry inlet reactant stream pressure (Pa), f_r is the reactant fraction in the dry inlet stream, R is the ideal gas constant (8.3143 J mol^{-1} K^{-1}), T is the temperature (K), i_e is the inlet reactant flow rate equivalent current density (A m^{-2}), and f is the inert gas to reactant fraction in the dry inlet stream.

Fig. 1 shows potentiostatic polarization curves (H$_2$/O$_2$+N$_2$) for the ten individual segments and the cell. All segments as well as the cell approach a limiting current at low cell voltages. The Fig. 1 inset presents model validation results using Eq. 1 for the determination of k. The model is accurate and leads to an O$_2$ mass transfer coefficient in N$_2$ media of 0.00862 m s^{-1}.

The mass transfer coefficient k is a series combination of mass transport resistances in gas k_g (m s^{-1}) and ionomer

electrolyte k_e (m s^{-1}) phases (1):

$$\frac{1}{k} = \frac{1}{k_g} + \frac{1}{k_e} \qquad [2]$$

Eq. 2 implies that the use of different gas diluents only affects k_g. Fig. 2 presents a strong correlation between the overall O$_2$ mass transfer coefficient and the diluent molecular weight. An extrapolation to a zero diluent weight gives the O$_2$ mass transfer coefficient in the electrolyte phase, which is equal to 0.0135 m s^{-1}. The application of the method for the characterization of MEAs with different Pt loadings and GDLs will be presented and discussed.
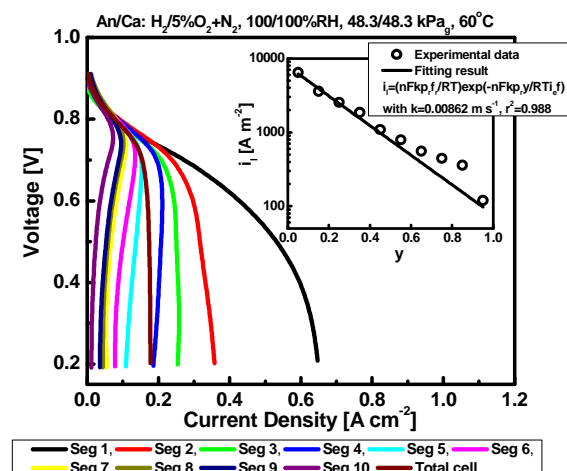

Fig. 1. Individual segment and overall polarization curves, and local limiting current density model validation (inset).
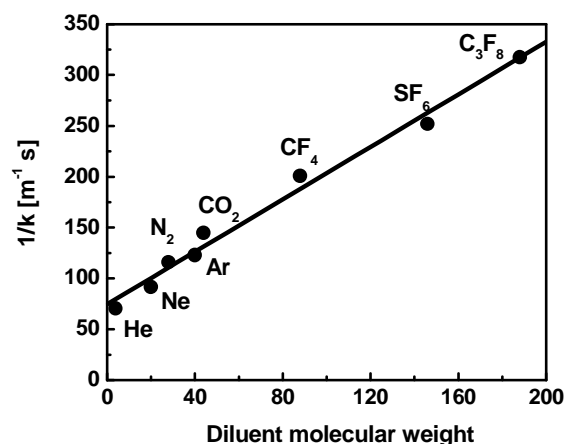

Fig. 2. Effect of the diluent molecular weight on the overall O$_2$ mass transfer coefficient.

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