Conf-921036--1

LA-UR- 92-1434

Title

COMPONENTS AND MATERIALS ISSUES IN POLYMER ELECTROLYTE FUEL CELLS FOR TRANSPORTATION APPLICATIONS

LA-UR--92-1434

DE92 013384

Author(s):

C. R. Derouin, MEE-11
T. E. Springer, MEE-11
F. A. Uribe, MEE-11
J. A. Valerio, MEE-11
M. S. Wilson, MEE-11
T. A. Zawodzinski, MEE-11
S. Gottesfeld, MEE-11

Submitted to.

Symposium on Batteries and Fuel Cells for Transportation
The Electrochemical Society
Toronto, Ontario, CANADA
October 11-16, 1992

Los Alamos
NATIONAL LABORATORY

Components and Materials Issues in Polymer Electrolyte Fuel Cells For Transportation Applicatiopns

C. Derouin, T. Springer, F. Uribe, J. Valerio, M. Wilson, T. Zawodzinski
and S. Gottesfeld

The Electronics Research Group
Los Alamos, NM 87545

Abstract

Recent research work on the polymer electrolyte fuel cell (PEFC) is described. This research work adresses the goal of bringing the PEFC technology to the performance and the cost levels required for its wide spread use in transportation. The main topics are (a) a new approach to the fabrication of Pt/C catalyst layers of high performance, employing loadings as low as 0.1 mgPt/cm^2; (b) measurements and modeling of membrane, cathode catalyst and cathode backing contributions to cell losses in the PEFC; and (c) carbon monoxide poisoning of anode electrocatalysts in the PEFC--the problem and possible solutions.

INTRODUCTION

The interest in the polymer electrolyte fuel cell(PEFC) as a potential power source for terrestrial transportation aplications has increased significantly in recent years. The basic features of high power density, perfect tolerance to CO_2, pseudo-solid-state character and low temperature of operation have been recently complemented by demonstrations of high performance on low Pt loadings and possible solutions to the problem of catalyst poisoning by CO. Whereas several demonstration projects have already started aiming at proving the feasibility of electric vehicle power systems based on PEFCs, questions such as water management optimization, factors determining cell longevity, increased reformate/air cell performance and the major question of the cost of this fuel cell technology are still waiting for complete answers. The main goal of the PEFC Core Technology Program at Los Alamos National Laboratory has been to conduct the basic and applied research necessary to bring the PEFC technology to the performance and cost levels required for its widespread use in transportation. The goals include (1) reducing the intrinsic cost, (2) increasing the energy and power density, (3) optimizing the system for operation on reformed organic fuels and air, and (4) achieving stable and efficient long-term operation. The central elements of the PEFC that need to be addressed to reach these goals are the loading and utilization of the Pt electrocatalyst, the proton and water transport in the ionomeric membrane electrolyte, the elimination of effects of poisoning of the anode electrocatalyst by traces of CO, and e mechanisms for long-term performance degradation and for cell failure. We :scribe in this contribution recent developments in our work on the above topics.

ELECTROCATALYSTS FOR THE PEFC

A NEW TECHNIQUE FOR IONOMERIC MEMBRANE CATALYZATION - We cently developed a new type of membrane-electrode assembly for our PEFCs[1]. We ıve achieved the highest catalyst utilization to date with this assembly, which is based ı a new structure for the catalyst layer and a new mode of bonding it to the ionomeric embrane.

The new electrode structure attempts to improve upon the construction based on ·ototech electrodes hot-pressed onto the membrane[2-5] by significantly increasing e contact area between the polymer electrolyte and the platinum clusters. This crease is achieved in two ways. First, the supported catalyst and the ionomeric lditive are cast together to form the catalytic layer, to assure that the thickness of the ıtalyst layer coincides with the depth of the ionomer. Second, the contact area :tween the ionomer additive and the catalyst is increased by completely eliminating e Teflon component and by improving the dispersion of the ionomer throughout the ıtalyst layer. The latter is accomplished by blending the solubilized ionomer and the atinized carbon into a homogeneous "ink" from which the thin film catalyst layer of e electrode is cast. The gas diffusion portion of the electrode consists of a separate :flonized carbon cloth backing that is stacked behind the thin catalyst film to provide ıpport and to provide a hydrophobic distribution network for the gases. This two-ırt construction of the electrode--hydrophobic backing and hydrophilic catalyst layer-llows each of the two regions to be fabricated separately with the properties that best ıit the function of each region. The new catalyst layer which is essentially a recast afion®-Pt/C composite, was initially prepared in the form of an ink that was coated ıto a Teflon film "blank."[1] The ink on the blank was baked in a forced convection ·en until dry. The coated blank and a polymer electrolyte membrane electrode were en hot-pressed together. The assembly was next removed from the press and cooled, en the Teflon blank peeled away leaving the thin film adhered to the membrane.[1]

This mode of fabrication is a return to the concept of a "catalyzed membrane." owever, in great contrast to the catalyzed membranes based on a layer of Pt black ixed with Teflon, as employed in the GE/Hamilton Standard technology, the thin talyst layer is now based on a supported Pt catalyst with very low Pt loading. ompared with the Prototech electrode/ionomer assemblies we employed extensively the last few years,[2-5] this new thin film technology has the advantage of a much inner catalyst layer (of the order of 3 to 5 microns) with a very uniform mix of talyst and ionomer. Some further modifications in the protocol of the fabrication of e catalyzed membrane have recently allowed us to apply the ink directly to the embrane (no blank required) and to obtain the results shown in Figures 1a and 1b ıth loadings of 0.17 and of 0.13 mgPt/cm^2/electrode, respectively The key to this ·w mode of membrane catalyzation has been to maintain both the membrane and e ionomeric component of the "ink" in the Na$^+$ form during the fabrication of the sembly, enabling higher temperatures of curing resulting in more robust films.[1b] ıe performance shown in Figure 1a was obtained with an ionomeric membrane pplied by Chlorine Engineers of Japan ,and designated by the manufacturer as embrane "C" The performance shown in Figure 1b was obtained with the XUS

13204.10. Dow membrane. The experimental Dow membrane in its fully humidified form is thinner than the fully humidified form of Membrane C (125 µm *vs* 200 µm). It is quite remarkable that such low Pt loadings can generate a current density above 3 A/cm^2 at a cell voltage higher than 0.4 V for operation on pressurized O_2, and, particularly, that such low loadings are sufficient to reach a cell voltage of 0.65 V at 1 A/cm^2 for cells operated on pressurized air. Figure 2 compares catalyst utilization for the three modes of membrane/catalyst assembly preparation: GE/Hamilton Standard (Pt black), Prototech electrode-based assemblies[2-5] (Pt/C), and the new thin film technology for membrane catalyzation. Catalyst utilization is significantly better with our new approach, particularly at the higher current densities (lower cell voltages). The latter aspect shows the advantage of thin catalyst layers in minimizing losses within the layer at higher current densities. This aspect, as well as other aspects of the performance of PEFCs at high current densities, is addressed in the new model we developed for the catalyst layer in these fuel cells, discussed in the next section.

A MODEL FOR THE CATHODE IN A PEFC - The ionomeric membrane separato has been the main focus of attention in performance evaluation and in modeling of PEFCs. However, in H_2/air PEFCs the losses originating from the air cathode are dominant at both low and high current densities. Following our previous work on ionomeric membrane transport properties and their modeling under PEFC operating conditions[6], we have extended our evaluation and modeling work to the detailed properties of the cathode catalyst layer[7] and the cathode backing This results in the complete one dimensional model for the PEFC, described schematically in figure 3. The model considers three components of the PEFC: the ionomeric membrane, the cathode catalyst layer and the cathode backing. (To first approximation anode losses can be negelected). Our model is based on detailed physicochemical characterization of each of these cell components, and is an effective tool for the evaluation of the performance of PEFCs tested in our laboratory. The iR-corrected polarization curves in figure 4 , recorded for a PEFC with the new type of thin-film catalyst layers, reveal that in the case of cell operation on neat O_2 most of the loss is from the membrane resistance (cell impedance measured at 5 kHz) , but in the case of operation on air the most significant contribution to cell losses comes from the cathode. This situation is enabled by the thinner and better ionomeric membranes demonstrated recently, which minimize cell resistance at high current densities Such advanced membranes leave the air cathode as the most significant source of cell losses in H_2/air PEFC's.

The "limiting current" in a fuel cell (see figure 4b) is usually ascribed to reactant transport limitations. However, the measured PEFC characteristics at high c.d.s do not agree with a simple model of a thin film barrier (ionomer or water) separating the reactant gas from the catalyst site. Such a simple model predicts a much steeper fall in cell voltage. Furthermore, if excess water around the cathode catalyst is the cause of the problem, the cell performance should have been rather unstable at the highest c.d.s, but this has not been the case. Also, Figure 4 shows that at high cell polarization the ratio between the iR corrected currents obtained on O_2 and on air of same total pressure is much smaller than 5:1, the ratio expected for a case of a thin film barrier We recently developed a model for the catalyst layer that appears to explain these haracteristic patterns[7] . The most important feature of our catalyst layer model is he detailed consideration of the counter fluxes of oxygen and protons. As a result of his fundamental property, described schematically in Figure 5, combined ermeability/conductivity limitations can arise within the catalyst layer at high urrent density. Such limitations are treated by this model.

The catalyst layer is envisioned in our model as being composed of four uperimposed and intimately cross-connected media:

(1) A diffusion medium that transports the reactant gas to the catalyst sites , both through tortuous, restricted channels in gaseous form and through the conducting polymer in dissolved form. This process is represented by a single effective diffusion coefficient D, and an effective oxygen concentration C for the hybrid medium.

(2) An ionically conducting medium with effective conductivity σ for hydrogen ions,

(3) An electronically conducting medium with no resistance, and

(4) A Pt catalyst, uniformly dispersed through the catalyst layer, with a rate constant k_0 (cm/s) for the oxygen reduction process at the open-cell potential, Tafel kinetics having a Tafel slope b in natural log units, overpotential η measured from the open cell potential, and effective surface area per unit volume A_V.

Reactant with a bulk concentration C^* is introduced at z=0 (the gas side of the catalyst ayer), and the protonic membrane interfaces with the catalyst layer at $z=\ell$. The roduction of current density I, the overpotential h, and the reactant concentration hrough the catalyst layer are described along z by the following three differential quations,

$$\frac{dI}{dz} = nFk_0A_vCe^{\eta/b}, \quad \frac{d\eta}{dz} = \frac{I}{\sigma}, \quad \frac{dC}{dz} = \frac{I - I_\ell}{nFD},$$

with the boundary conditions $I(0)=0$, $\eta(0)=\eta_0$, and $C(0)=C^*$ (I_ℓ is the total integrated urrent density).

Figure 6 shows a profile of the variables η/b, I/I_ℓ, C/C^* and $d(I/I_\ell)/d(z/\ell)$ as a unction of z/ℓ for an overpotential that is high enough to drop the reactant oncentration to zero within the catalyst layer. Under such circumstances, and if the rotonic conductivity within the layer is insufficient, a loss is caused by the equirement that protons reach the active domain of the catalyst layer which is now ocated near the gas boundary and away from the membrane. This situation is redicted to bring about a "soft" current limit , with a discernible finite increase in lope of the polarization curve. The iR-corrected curves in figure 4 b show that the neasured current limit is indeed soft, as predicted by the model. We can now nderstand this characteristic form of fall in cell voltage as the result of the limited nobilities of oxygen and protons within the catalyst layer. It is this mixed ermeability/conductivity control that lowers the ratio of currents measured at high cell polarizations on 5 atm O_2 vs. 5 atm air. This is expected when the cell current is controlled in part by a conductivity factor which does not depend on reactant concentration Figure 7 shows calculated effects of the magnitudes of the effective diffusion coeffecient of O_2 and the effective protonic resitivity within the cathode catalyst layer. It is clear from this figure that the transport characteristics within the catalyst layer have a profound effect on the performance of air cathodes in PEFCs. One key property of the catalyst layer is its overall thickness. The smaller the thickness the smaller will be losses originating from combined permeability/conductivity limitations, as can be understood from examination of the curves in figure 6. Our newest PEFC catalyst layers[1] are 3-5 μm thick, as compared with 50-100 μm thick catalyst layers emplyed in previous PEFCs with low Pt laodings [2-5]. As a result, the penalties for lower permeabilities at low oxygen partial pressures are minimized, enabling good performance even at 1 atm of air. This is demonstrated in Figure 8 which shows experimental polarization curves recorded for PEFCs with the new thin-film catalyst layers in the pressure range 1-5 atm. for both neat O_2 and air.

Figure 7 also contains the experimental manifestation of the significant contribution of the cathode backing to cell losses.This is seen from the better high-current performance of a PEFC operating on1 atm O_2 vs. the same PEFC operating on 5 atm air. Such a difference between cathodes operated at the same partial pressure of O_2 but at different total gas pressure is expected only in the presence of a significant gas-phase diffusion barrier, which must be located in the backing of the cathode. The model we use for the cathode backing assumes no pressure drop within this layer. The change in mole fraction of the gaseous components through the backing is described by the Stefan-Maxwell equation., assumed to hold for the three components N_2, O_2 , and water vapor. When water vapor saturation takes place in the backing, the water mole fractiion in the gas phase will remain constant and water flux will be only in the liquid phase.We assume no blocking effects caused by liquid water in the backing. A demonstartion of calculated losses expected from the cathode backing are shown in figures 9 and 10. Figure 9 shows the calculated (iR-corrected) performance of a PEFC with a given catalyst layer in three cases : no backing losses, backing losses at1 atm O_2 , and backing losses at 5 atm air The backing layer assumed in each case is 300 μm thick and has 40% porosity, and the temperature assumed is 80°C. The larger losses caused by gas transport through the backing at 5 atm air explain the relative performances obtained in PEFCs on 5 atm air vs. 1 atm O_2 (see Fig. 8). Figure 10 shows that the effect of the backing on cell performance is, in fact, smaller in the case of 1 atm air than in the case of 5 atm air. This is an expected result (less collisions of O_2 molecules with N_2 molecules on their way to the catalyst at lower overall pressure) which means that a further increase in performance of PEFCs at 1 atm air may probably come primarily from improvements in catalyst layer properties rather than properties of the backing. An important aspect which needs to be remembered, however, is the possibility of partial "flooding" of the backing. To the extent that this latter effect, which has been neglected in our model, becomes significant, clogging of the porous gas filled network within the backing can take place, and the transport losses within this layer may become much more pronounced. This situation is likely to occur only in

:her ill-designed or aging backing layers which have lost their hydrophobic aracteristics.

ıFE-TESTING OF SINGLE PEFCs WITH LOW PLATINUM LOADINGS

new set of fuel cell testing stations installed recently in our laboratory has allowed to run life tests of PEFCs based on low Pt loadings under conditions of continuous essurization and elevated cell temperatures. We report here results of recent life ;ts of PEFCs based on various types of ionomeric membranes. *All of these tests monstrated that PEFCs based on Pt loadings as low as 0.1 mgPt/cm^2 can operate ntinuously for several thousand hours showing only limited gradual loss of rformance.*.This gradual loss of performance is typically of the following nature. If itial cell voltage (on 3 atm H_2 and 5 atm air) @ 1 A/cm^2 is , typically, 0.64V, the ltage at the same current density will fall to 0.60V after a week of continuous cell eration, and further to 0.53V after a full month of continuous operation. Out of the erall loss in cell voltage @ 1A/cm^2, only about 20% is caused by a gradual increase (high frequency) cell resistance, whereas the rest has to do with some terioration of the permeability/conductivity characteristics of the catalyst layer, d/or with partial loss of hydrophobicity in the backing of the cathode.

everal life tests were performed by us on PEFCs with membranes.thinner than ual. (50 μm vs. the usual thickness of 100-200 μm). Such thin membranes are tractive from the point of view of cell performance (see next section) but they ise questions of integrity and, thus, of cell longevity. In the past we found it fficult to operate cells based on such thin membranes for prolonged periods cause of membrane puncturing problems. This was particularly severe for cells in hich the cathodes operated on pressurized neat O_2 To solve this problem in cells sembled by hot pressing catalyzed carbon electrodes to ionomeric membranes, we otected the area of the membrane around the edge of the electrode by a well-signed gasket. This mode of protection of what seems to be a sensitive part of the nomeric membrane in the cell, has resulted in a very substantial improvement in e longevity of PEFCs based on thin ionomeric membranes. For the most recent type catalyzed membranes demonstrated by us [1b] , hot-pressing is not required at all complete the assembly of the PEFC, and, therefore, the problem of membrane inning around the edge of the electrode is relieved. .This further improves the tegrity of thinner membranes. A life test of a PEFC with a 50-μm-thick Nafion® W =1100) membrane (hot-pressed assembly with electrdoe edge protection) was n in our testing facility for 60 consecutive days with no apparent damage to the in membrane. During this time the cathode operated on pressurized O_2 during ytime and on pressurized air during nighttime. (The anode feed stream was neat $_2$)..A recent life-test of a PEFC with another experimental membrane of similar ickness has also demonstrated long-term membrane integrity. The conclusion om these results are (a) significant improvements in the longevity of PEFCs can be hieved by identifying problem areas in the membrane/electrode assembly and ressing them , and (b) PEFCs that have thinner ionomeric membranes could be ore practical than was previously believed. This is important because such thin embranes enable high performance thanks to their superior conductivities and ater transport characteristics (see next Section).

WATER TRANSPORT AND THE STEADY-STATE WATER PROFILE IN THE IONOMERIC MEMBRANE OF AN OPERATING PEFC

The issue of water management is very central in the successful long-term operation of a PEFC. Spatial variations of water content within the polymeric electrolyte of a current-carrying fuel cell result from the electroosmotic dragging of water with proton transport from anode to cathode, the production of water by the oxygen reduction reaction at the cathode, humidification conditions of the inlet gas stream, and "back-diffusion" of water from cathode to anode. We recently developed an isothermal one-dimensional, steady-state model for water transport through a PEFC.[6] The model includes transport of water through the porous electrodes, based on calculated diffusivities corrected for porosity, and transport throughout the membrane electrolyte that is evaluated on the basis of experimentally determined parameters. The added detailed experimental characterization is an important feature of our modeling effort for water transport in the ionomeric membrane: we employed water sorption isotherms, water diffusion coefficients, electroosmotic drag coefficients, and membrane protonic conductivities, all of which have been *measured* in our laboratory as a function of membrane water content.[6,8] .We deemed it highly desirable to use, in our model, a complete set of experimental data generated under conditions in which the membrane is routinely handled and tested for PEFC work, i.e., conditions of partial and variable hydration with no added liquid electrolyte. We evaluated the intradiffusion coefficient of water in the membrane by Pulsed Field Gradient Spin Echo (PGSE) NMR.[9] For the Nafion® 117 membrane we found that the water intradiffusion coefficient varies between $6x10^{-6}$ cm^2/s and $6x10^{-7} cm^2/s$ as the degree of hydration falls from 14 to 2 H_2O/SO_3H[9]. The electroosmotic drag in the same Nafion® 117 membrane seems to fall with water content, from 2.5 H_2O/H^+ at full membrane hydration to negligible water drag at very low water content [8]. On the basis of these experimentally evaluated water-diffusion coefficients and water drag as functions of membrane water content, and of boundary conditions that apply under PEFC conditions, a steady-state profile of water in the membrane could be solved for a given set of external humidification conditions. An example of the results of such calculations is given in Figure 11, which shows calculated profiles of water in the Nafion® 117 membrane at different current densities under ordinary cell humidification conditions at 80°C. It is clear that a significant lowering in water level is expected under these conditions near the anode. Figure 12 contains some computed curves evaluated from the calculated water profiles at steady state and the measured dependence of membrane conductivity on water content. The upper part of the figure shows calculated cell resistance, (i.e., membrane resistance) as function of cell current. The lower part shows the calculated ratio of net water flux / proton flux through an operating PEFC. The upper part of this figure demonstrates clearly the advantage of thinner membranes in achieving high PEFC performance. The advantage of lowering membrane thickness is seen to be nonlinear, i.e., above and beyond the linear increase in electrolyte conductivity expected from membrane thickness ratios. This nonlinear effect is caused by the more effective back diffusion of water from cathode to anode in thinner membranes, which ensures a high level of hydration throughout the membrane, even at high current densities. We confirmed during our PEFC testing experiments that thin membranes indeed have strong advantages in achieving high cell performances. Obviously, a complete comparative evaluation of ionomeric membranes for PEFCs should depend on samples of equal thickness, as well as on the tradeoffs between cell performance and longevity. The lower part of Figure 12 shows that the net water flux through the operating PEFC is expected to be significantly smaller than the measured drag of water in fully hydrated ionomeric membranes, as we have confirmed experimentally[8]. This is the result of the steep profile of water in the operating cell, as shown in Figure 11. Again, thinner membranes are associated with a minimized net flux of water, an additional advantage in the design of a practical system.

Finally, we have shown recently in a study of the kinetics of oxygen reduction at the Pt/recast ionomer interface [10], that a high level of hydration is required at the Pt/ionomer interface to enable high rates of oxygen reduction This beneficial role of water is understood to originate from facilitated proton transfer and from minimized interactions of anionic as well as hydrophobic components of the ionomer with the Pt catalyst surface[10].

ELIMINATION OF CO POISONING EFFECTS AT THE ANODE ELECTROCATALYST

Pt electrocatalyst poisoning by traces of CO is very severe at 80°C. Levels of CO as low as 10 ppm have a significant effect on PEFC performance.[11] This is true for cells with lower, as well as higher, Pt loadings. We reported on a novel approach to the elimination of such effects by bleeding low levels of O_2 into the CO-contaminated H_2 feed stream.[11,12] In recent tests we established that this technique is very effective under realistic conditions of unity stoichiometric flow of H_2. In fact, the level of O_2 required for complete cleansing of the anode electrocatalyst is lower under conditions of low H_2 flow, requiring only 2% of air to eliminate poisoning effects of 100 ppm CO.[13] Analysis of anode exhaust gases for PEFC anodes fed with $H_2/CO/O_2$ mixtures has shown that CO is completely eliminated from the gas mixture when the anode feed stream levels are 100 ppm CO and 0.4% air.[13]

CONCLUSIONS

(1) Polymer electrolyte fuel cells can operate on very low Pt loadings (0.1 mg/cm^2) while achieving high performance. There seems to be only a limited gradual deterioration in performance of such cells in tests lasting for 2000 hours.

(2) Limiting currents in PEFCs with air cathodes apparently result from combined permeability/conductivity limitations in the cathode catalyst layer. Increased

performance of PEFCs, particularly at low air pressures, can be most probably achieved by further optimization of catalyst layer composition and structure.

3) Modeling of steady-state water profiles and resistive losses in ionomeric membranes in operating PEFCs reveal the advantage of thinner membranes in preventing significant water losses near the anode at high current densities. These advantages of thinner membranes have been verified experimentally. Longevity of some PEFCs based on thin (50 μm) ionomeric membranes has been demonstrated.

4) Elimination of CO poisoning effects at the anode electrocatalyst by continuous bleeding of low levels of air seems to be a viable approach when operating PEFCs on reformed methanol.

REFERENCES

1]. (a) M. S. Wilson and S. Gottesfeld, *J. Appl. Electrochem.*, 22, 1 (1992); (b) *J. Electrochem. Soc.* 139, L28 (1992).

2]. I. D. Raistrick, J. W. Van Zee, R. E. White, K. Kinoshita and H. S. Burney (Eds.), in "Proceedings of the Symposium on Diaphragms, Separators and Ion Exchange Membranes, The Electrochemical Society" (1986), p. 172.

3]. I. D. Raistrick, U.S. Patent No. 4,876,115 (1989).

4]. E. A. Ticianelli, C. R. Derouin, and S. Srinivasan, *J. Electroanal. Chem.* 251, 275 (1988).

5]. E. A. Ticianelli, C. R. Derouin, A. Redondo, and S. Srinivasan *J. Electrochem. Soc.* 135, 2209 (1988) .

6]. T. E. Springer, T. A. Zawodzinski, and S. Gottesfeld, *J.Electrochem. Soc.*, 138, 2334 (1991).

7] T.E. Springer and S. Gottesfeld, in "Modeling of Batteries and Fuel Cells", R.E.White, M.E. Verbrugge ard J.F. Stockel Eds., Proceedings volume 91-10, The Electrochemical Society, 1991 pp 197-208.

8] T. Zawodzinski, T.E. Springer, J. Davey, J. Valerio and S. Gottesfeld, ibid, pp.187 196.

9]. T. A. Zawodzinski, M. Neeman, L. D. Silleiud, and S. Gottesfeld, *J. Phys. Chem.*, 95, 6040 (1991).

10] F. A. Uribe, T. E. Springer, and S. Gottesfeld, "A Microelectrode Study of Oxygen Reduction at the Platinum - Recast Nafion Film Interface," *J. Electrochem. Soc.*, 139,765 (1992).

11]. S. Gottesfeld and J. Pafford, *J. Electrochem. Soc.*, 135, 2651 (1988) .

12]. S. Gottesfeld, U.S. Patent No. 4,910,099 (1990).

13]. H. Tobias, M. T. Paffett, P. Pappin, J. Valerio, and S. Gottesfeld, in "Proceedings of the Direct Methanol/Air Fuel Cell Workshop," The Electrochemical Society, 1992.

cknowledgement

This work is supported by U. S. Dept. of Energy, office of Energy Conservation nd Renewable Energy.

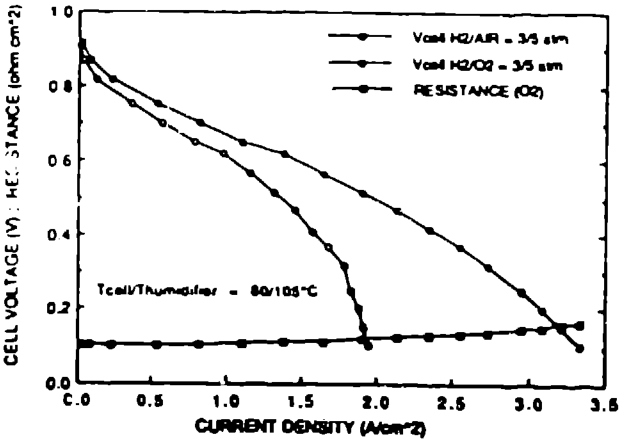


Figure 1a. Performance of a PEFC with a thin catalyst layer based on Pt/C catalyst bonded to the membrane. Loading - 0.17 mgPt/cm^2/electrode. The ionomeric membrane used here was membrane "C" (Chlorine Engineers, Japan).

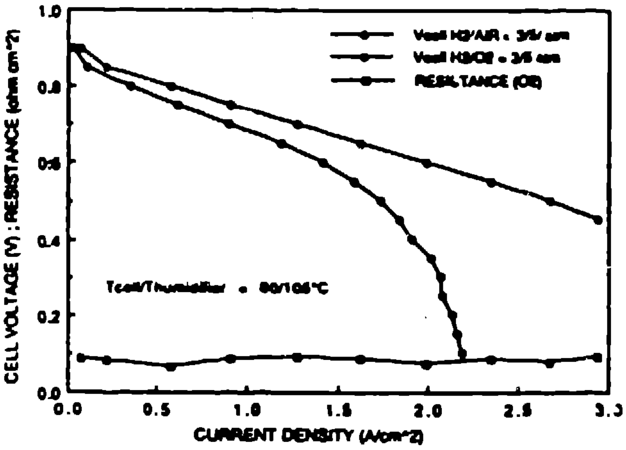


Figure 1b. Same as figure 1a, for Pt loading of 0.13 mgPt/cm^2/electrode, and an experimental Dow membrane.

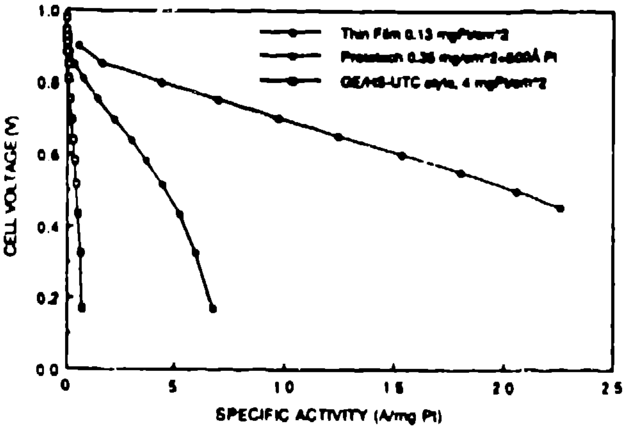
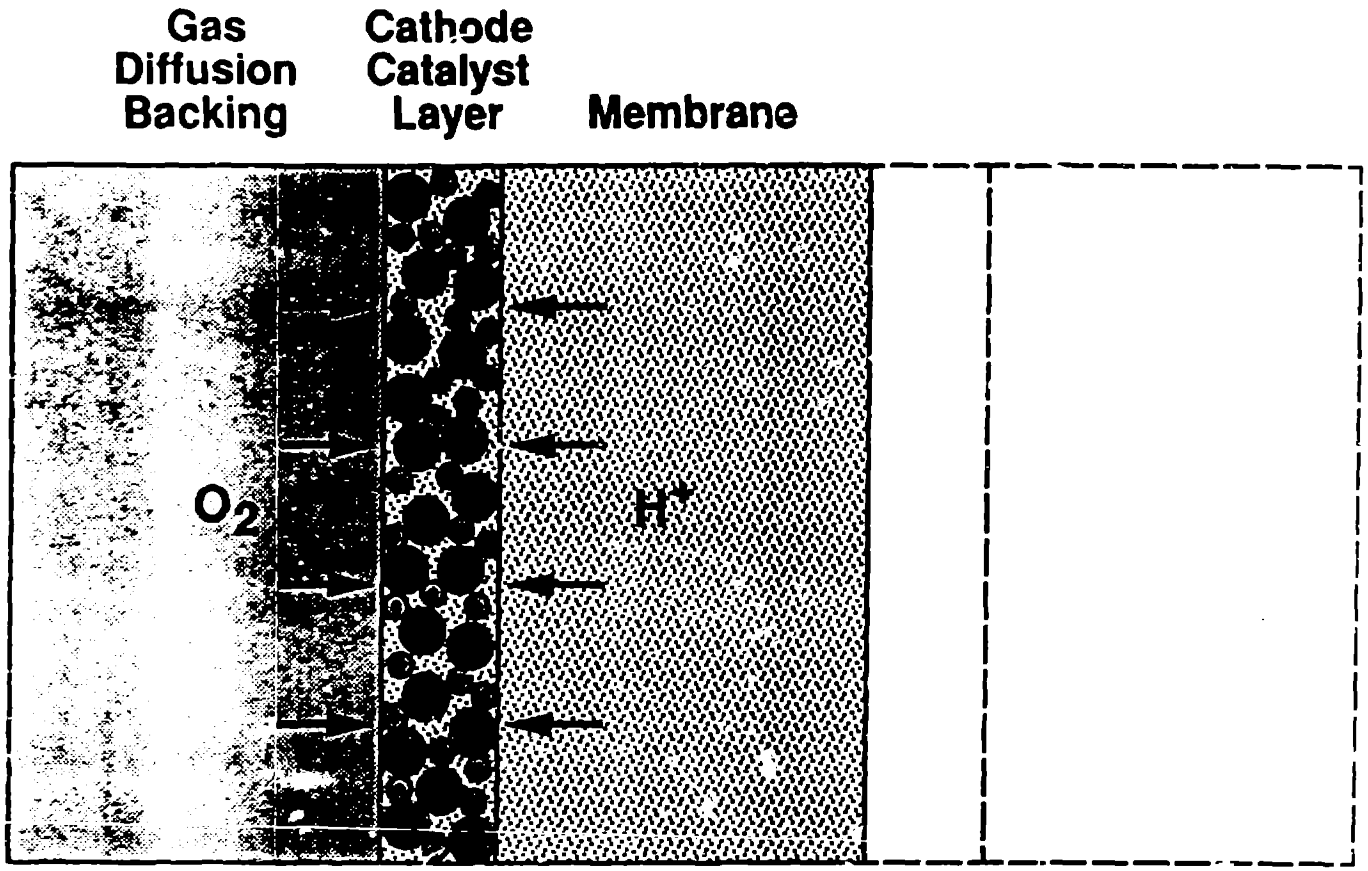


Figure 2. Comparison of specific Pt catalyst activities in PEFC's with three different types of Pt catalyst layers

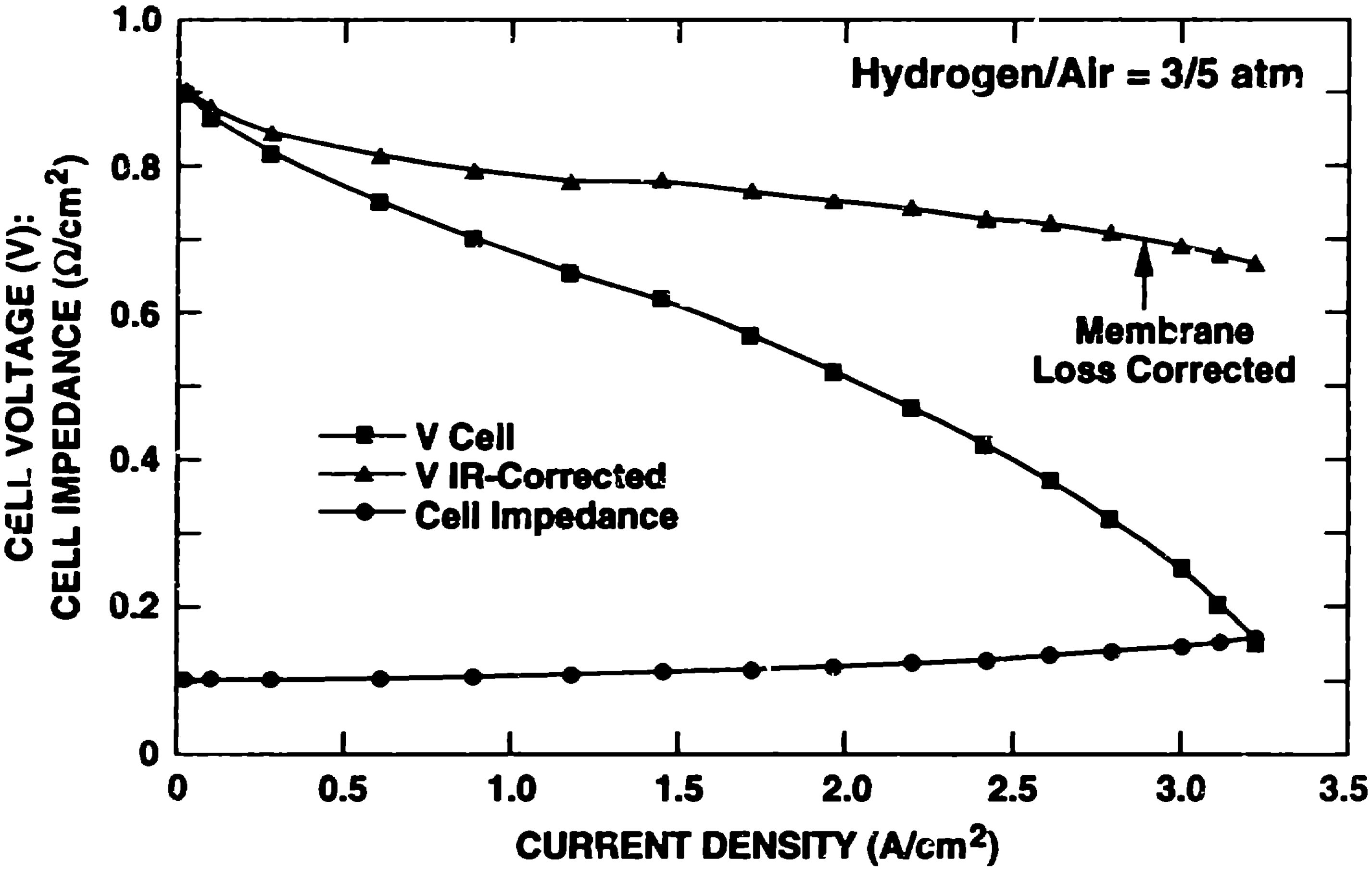
Three Components in the Complete Fuel Cell Model

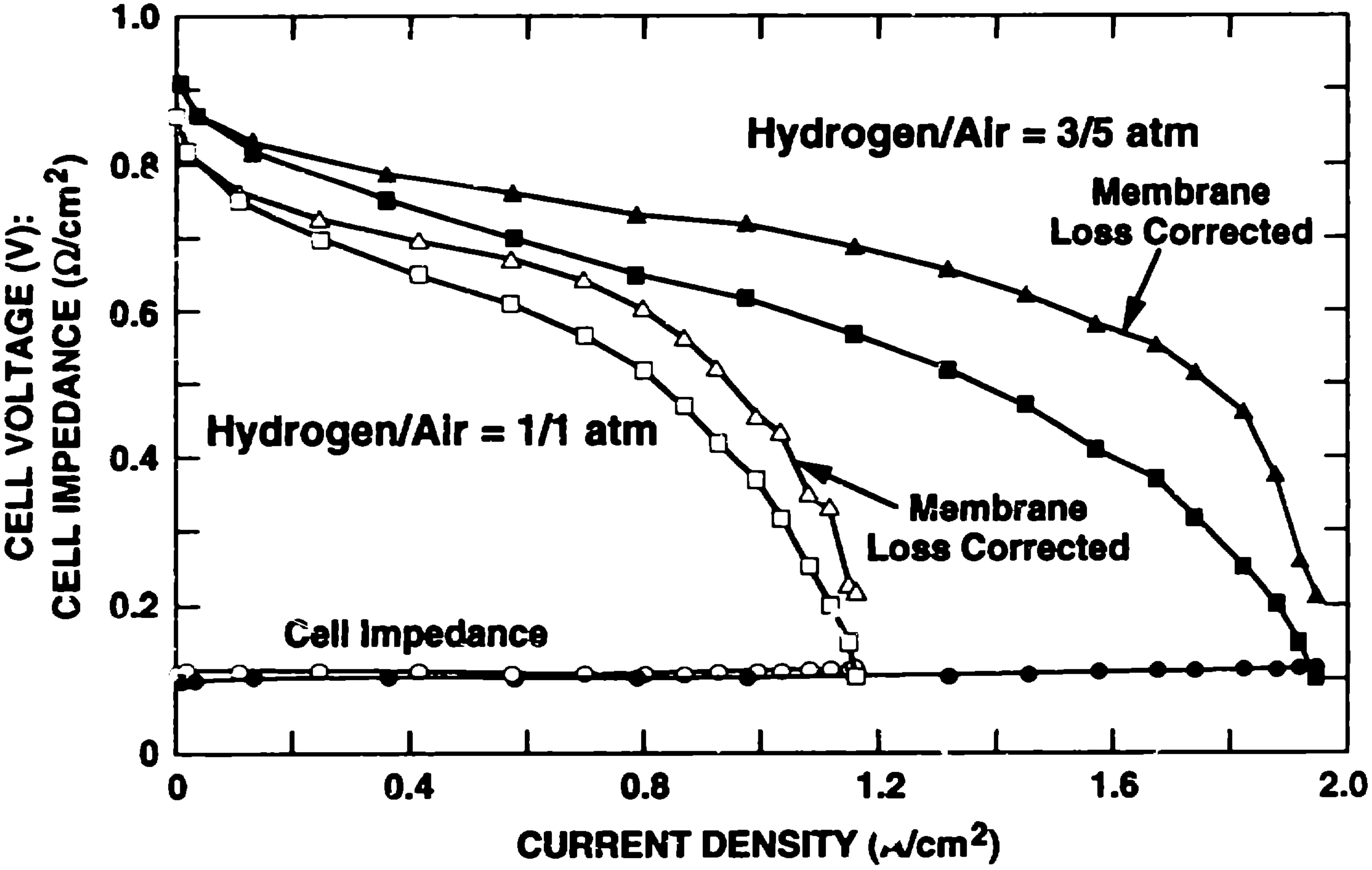


Gas Diffusion Backing

Cathode Catalyst Layer

Membrane


Hydrogen/Air = 3/5 atm
Membrane Loss Corrected
V Cell
V IR-Corrected
Cell Impedance
CELL VOLTAGE (V):
CELL IMPEDANCE (Ω/cm²)
CURRENT DENSITY (A/cm²)
1.0
0.8
0.6
0.4
0.2
0
0
0.5
1.0
1.5
2.0
2.5
3.0
3.5


Hydrogen/Air = 3/5 atm
Membrane Loss Corrected
Hydrogen/Air = 1/1 atm
Membrane Loss Corrected
Cell Impedance
CELL VOLTAGE (V): CELL IMPEDANCE (Ω/cm^2)
CURRENT DENSITY (A/cm^2)
1.0
0.8
0.6
0.4
0.2
0
0
0.4
0.8
1.2
1.6
2.0

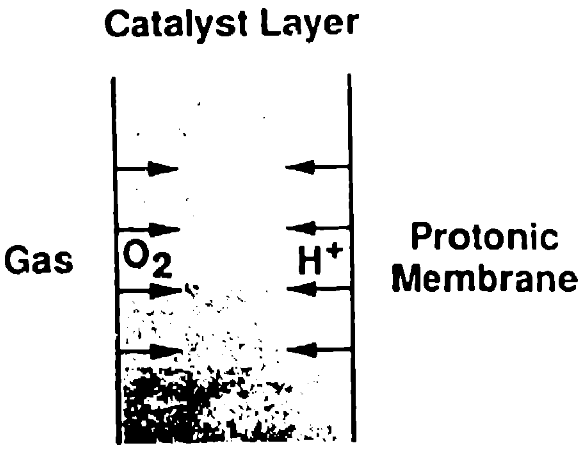


Figure 5. Schematic presentation of the counter fluxes of O_2 and of protons in a cathode catalyst layer considered in the modeling of this layer.

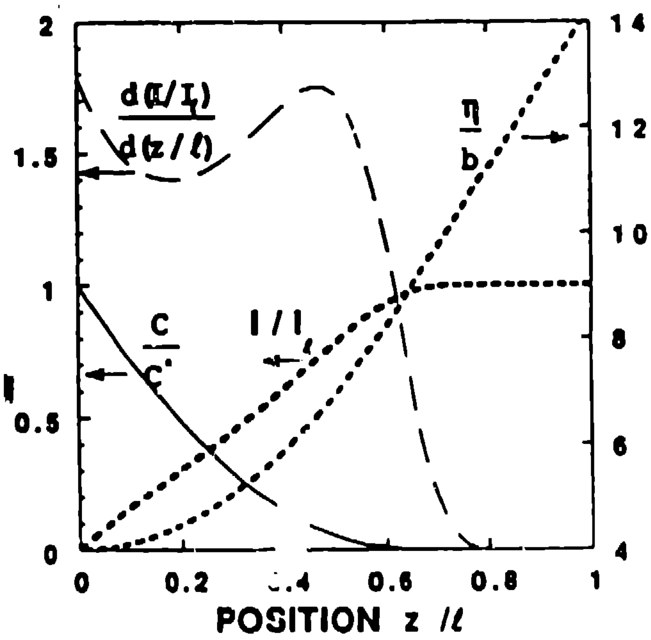


Figure 6. Illustrative profiles of integrated current density, overpotential, normalized oxygen concentration and current production through the catalyst layer.

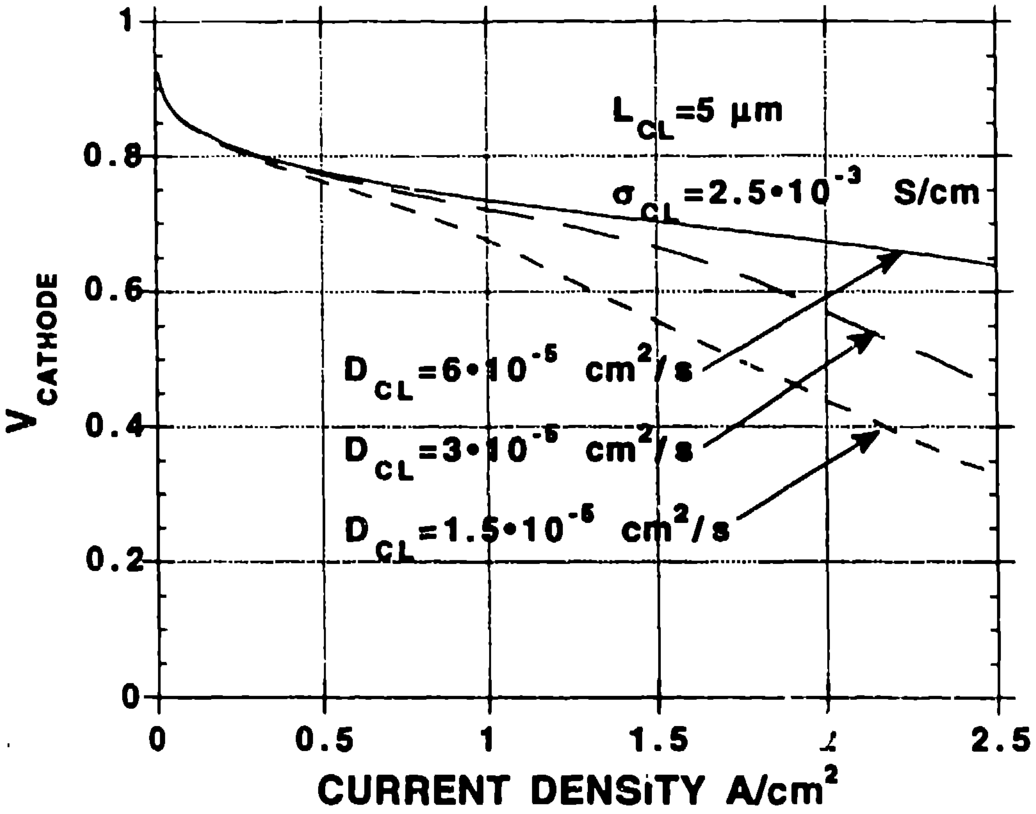


Fig. 7a. Calculated performance of catalyst layer without backing for three values of the effective diffusion coefficient.

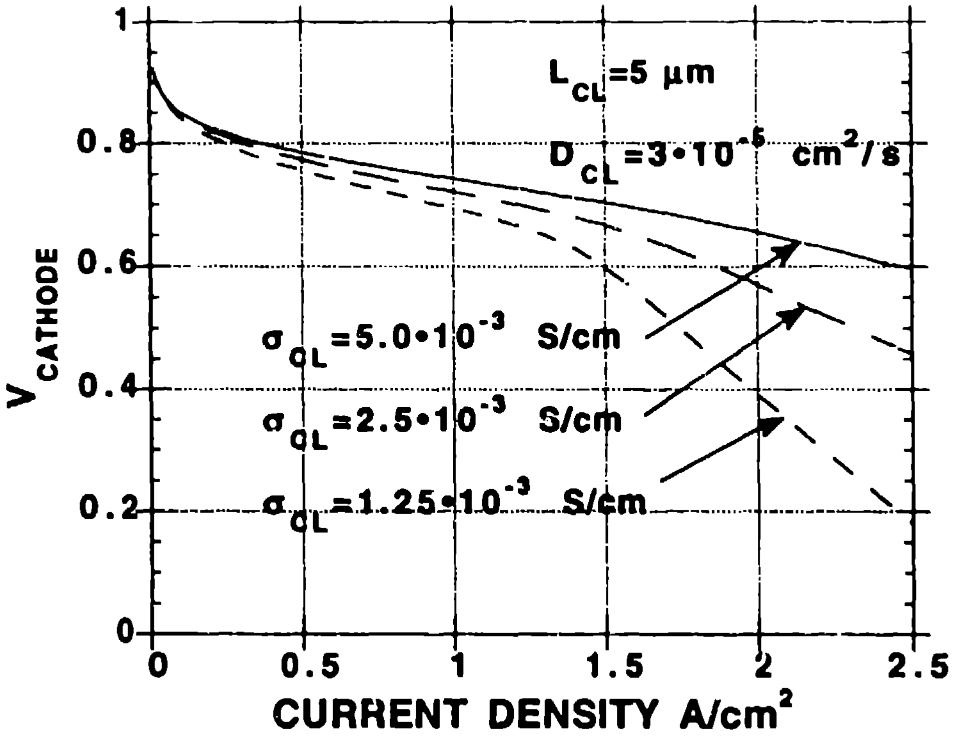
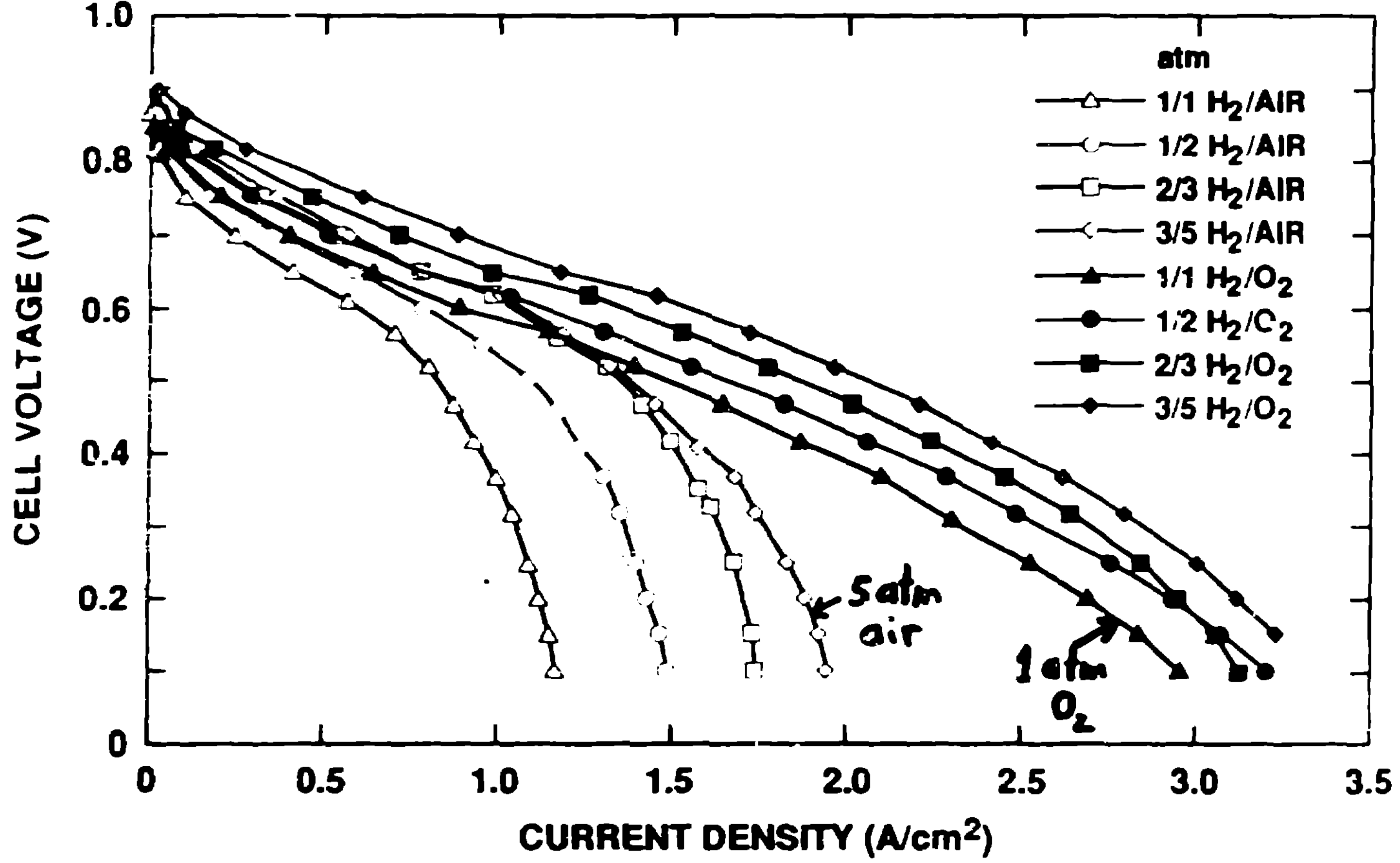


Fig. 7b. Calculated performance of catalyst layer without backing for three values of ionic conductivity.

Polarization Curves Illustrating Pressure/Gas Composition Dependencies of 0.17 mg Pt/cm^2 Catalyzed Membrane "C"



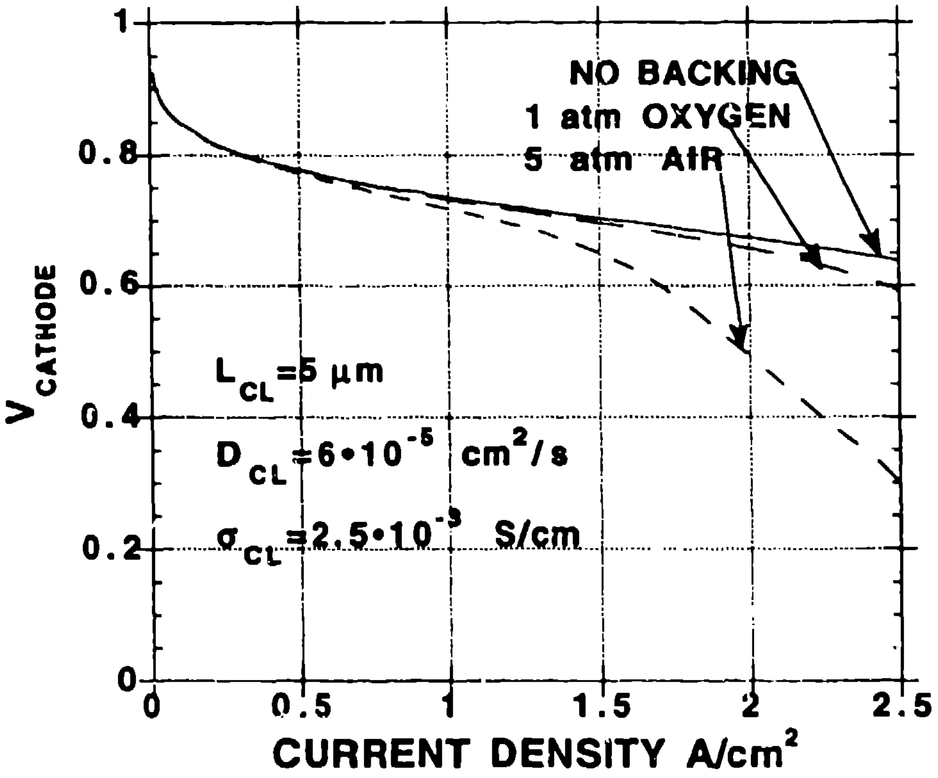


Fig. 9. The effect of a 300 μm backing is illustrated by comparing 1-atm O_2 and 5-atm air predicted performance. With no backing both cases are the same.

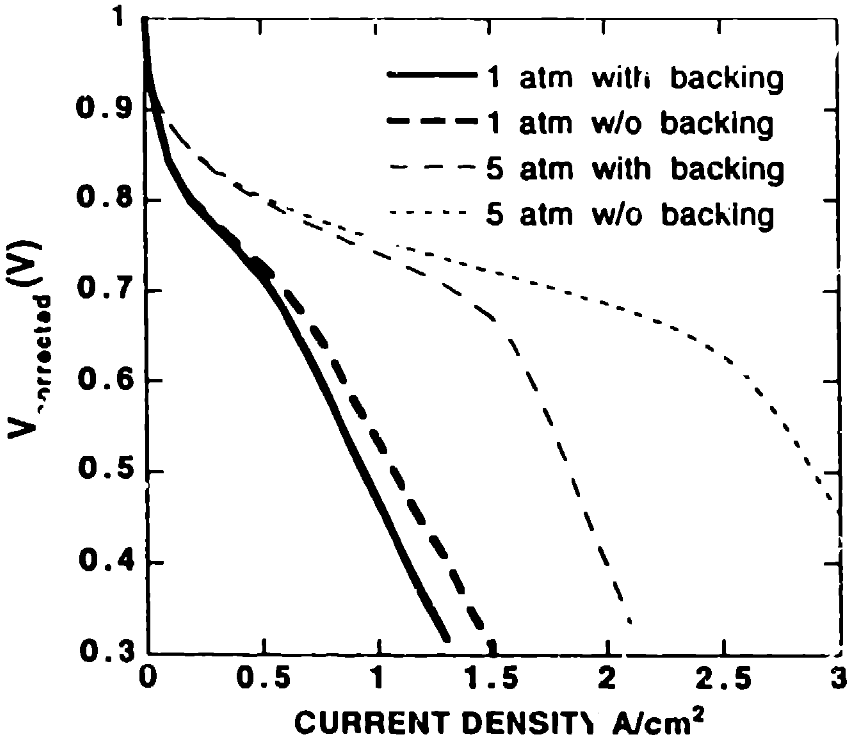

1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0
0.5
1
1.5
2
2.5
3
1 atm with backing
1 atm w/o backing
5 atm with backing
5 atm w/o backing
$V_{corrected}$ (V)
CURRENT DENSITY A/cm^2

FIGURE 10

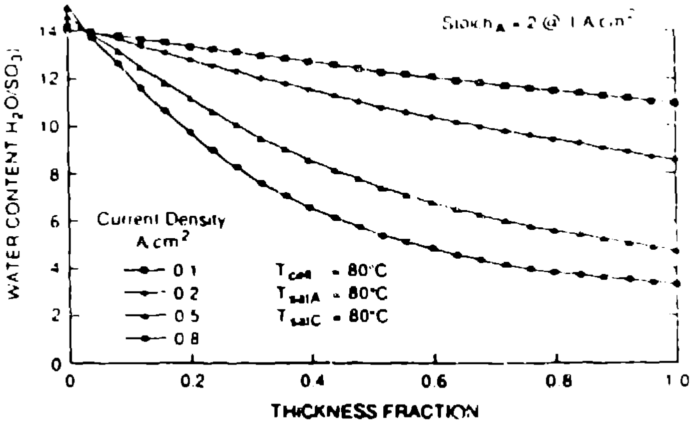


Figure 11. Computed water profiles in Nafion 117 for a cell operating at 80°C with water saturators at the same temperature. The cathode is on the left.

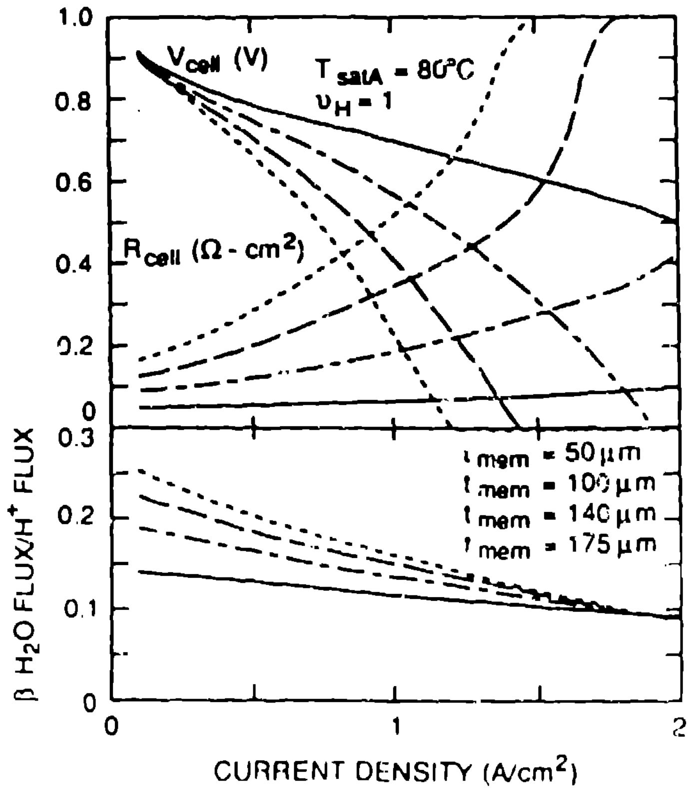


Figure 12. Computed V_{cell} and R_{coil} (upper part) and H_2O/H^+ flux ratio (lower part) for Nafion 117 membrane of different thicknesses. Cell and saturator temperature 80°C