Polymer electrolyte membrane (PEM) fuel cells are electrochemical devices capable of continuous conversion of the chemical energy of reactants into electrical energy. They are currently in development for a wide range of commercial applications and are especially important for stationary power sources.

Impedance measurements of PEM fuel cells provide the opportunity for in situ identification and quantification of physical phenomena which influence cell performance, but the small value of cell impedance complicates measurement. Impedance spectra typically exhibit inductive features at high frequency, and some authors report inductive loops at low frequencies. The high-frequency inductive features are understood to be caused by instrument artifacts, but the interpretation of the low-frequency inductive loops is less clear.

While the low-frequency loops have been tentatively attributed to side reactions, they could also be caused or influenced by nonstationary phenomena. The objective of this work was to use the measurement-model concept\(^1\) to assess the error structure of the impedance measurements taken for a PEM fuel cell.

**Impedance measurements of fuel cells.**—Impedance spectroscopy has been used extensively to study the behavior of PEM fuel cells. Merida et al.\(^2\) and Le Canut et al.\(^3\) have described the use of impedance spectroscopy for fuel cell diagnostics. Paganin et al.\(^4\) used impedance spectroscopy to study the effect of cell temperature, oxygen partial pressure, electrode composition, and membrane thickness on PEMFC performance. Andreaus et al.\(^5\) used impedance data obtained under high current densities to estimate performance losses in the fuel cell. Cha et al.\(^6\) studied flooding of a microflow channel. Lee et al.\(^7\) used impedance to evaluate the optimum Nafion loading in the catalyst active layer (CAL). Song et al.\(^8\) used impedance to evaluate the optimal composition of the catalyst-layer support material. They reported two capacitive loops for the impedance response of the fuel cell. Abe et al.\(^9\) used impedance to analyze the effect of humidity in the oxygen stream at the cathode of the PEM fuel cell. Impedance investigations have also been reported for different fuel cell applications such as studies of the effect of membrane thickness on the conductivity of the Nafion, characterization of electrospayed Nafion films, performance evaluation of self-humidified composite membranes, and characterization of single-walled carbon-nanotube-based proton exchange membrane assemblies for hydrogen fuel cells.

Cirreanu et al.\(^10\) reported impedance spectra with two loops. The high-frequency loop was attributed to the effect of reaction kinetics and oxygen transport in the CAL, and the low-frequency loop was attributed to mass-transport limitations of gases and water in the gas diffusion layer (GDL). The group also suggested a circuit analog model to explain the experimental results and calculated the parameters such as surface exchange current density and Tafel slopes. Many authors, e.g., Castañeda et al.\(^11\) and Li et al.,\(^12\) reported only one capacitive loop.

A recent impedance study reported by Makharia et al.\(^13\) revealed a capacitive loop at intermediate frequencies and an inductive loop at low frequencies. The capacitive loop was attributed to the response of electrochemical reactions occurring in the fuel cell, and the inductive loops were tentatively attributed to side reactions and relaxation of associated reaction intermediates. Their interpretation is consistent with that suggested by Antoine et al.,\(^14\) who proposed the presence of unspecified reaction intermediates. They suggested that low-frequency inductive loops were a result of the relaxation of adsorbed species involved in different steps of the oxygen reduction reaction. More recently, Siegel et al.\(^15\) considered a two-step hydrogen oxidation reaction and reported low-frequency inductive loops. They have explained that the inductive loops were the result of changing different factors such as water concentration, membrane thickness, hydrogen pressure, and the hydrogen oxidation kinetics. The influence of carbon monoxide poisoning on platinum and platinum-ruthenium anodes was investigated using impedance spectroscopy.\(^16\) The reported impedance response exhibited low-frequency pseudo-inductive behavior which was attributed to a surface relaxation process of competitive oxidation of hydrogen and carbon monoxide at the anode.

The inductive loops reported in the literature are typically seen at very low frequencies, e.g., 1 mHz, at which system stationarity must be questioned. The objective of this work was to use the measurement-model developed by Agarwal et al.\(^17\) to determine whether the low-frequency inductive loops were due to or influenced by nonstationary behavior.

**Measurement model.**—As described by Orazi,\(^18\) the measurement model was introduced as a means to resolve recurring issues in regression of impedance data, e.g.,\(^19\) (i) identification of the most appropriate weighting strategy for regression, (ii) assessment of the noise level in the measurement, and (iii) identification of the frequency range unaffected by instrumental artifacts or nonstationary behavior. The errors in an impedance measurement can be expressed in terms of the difference between the observed value \(Z(\omega)\) and a model value \(\hat{Z}(\omega)\) as

\[
e_{\text{res}}(\omega) = Z(\omega) - \hat{Z}(\omega) = e_{\text{err}}(\omega) + e_{\text{bias}}(\omega) + e_{\text{stoch}}(\omega) \quad [1]
\]

where \(e_{\text{res}}\) represents the residual error, \(e_{\text{err}}(\omega)\) is the systematic error that can be attributed to inadequacies of the model, \(e_{\text{bias}}(\omega)\) represents the systematic experimental bias error that cannot be attributed to model inadequacies, and \(e_{\text{stoch}}(\omega)\) is the stochastic error with expectation \(E[e_{\text{stoch}}(\omega)] = 0\).

A distinction is drawn, following Agarwal et al.,\(^17\) between stochastic errors that are randomly distributed about a mean value of zero, errors caused by the lack of fit of a model, and experimental
bias errors that are propagated through the model. The experimental bias errors, assumed to be those that cause a lack of consistency with the Kramers–Kronig relations, may be caused by nonstationarity or by instrumental artifacts. The problem of interpretation of impedance data is therefore defined to consist of two parts, one of identification of experimental errors, which includes assessment of consistency with the Kramers–Kronig relations, and one of fitting, which entails model identification, selection of weighting strategies, and examination of residual errors. The error analysis provides information that can be incorporated into regression of process models.

The measurement-model method for distinguishing between bias and stochastic errors is based on using a generalized model as a filter for nonreliability of impedance data. The measurement model is composed of a superposition of line-shapes which can be arbitrarily chosen subject to the constraint that the model satisfies the Kramers–Kronig relations. The model composed of Voigt elements in series with a solution resistance, i.e.,

$$Z = R_0 + \sum_{k=1}^{K} \frac{R_k}{1 + j\omega \tau_k}$$

has been shown to be a useful measurement model. With a sufficient number of parameters, the Voigt model was able to provide a statistically significant fit to a broad variety of impedance spectra.

The measurement model is used first to filter lack of replication of repeated impedance scans. The statistics of the residual errors yields an estimate for the variance (or standard deviation) of stochastic measurement errors. This experimentally determined variance is then used to weight subsequent regression of the measurement model to determine consistency with the Kramers–Kronig relations. If the data can be represented by a model that is itself consistent with the Kramers–Kronig relations, the data can be considered to be consistent. The concept of using a generalized measurement model to assess consistency with the Kramers–Kronig relations, first introduced by Agarwal et al., was also employed by Boukamp and Macdonald and by Boukamp using weighting strategies based on an assumed error structure. The experimental determination of the stochastic error structure as used here, however, allows formal quantification of the extent of agreement with the Kramers–Kronig relations.

Other transfer-function models can be used as a measurement model so long as they are consistent with the Kramers–Kronig relations. Shukla and Orazem have demonstrated that the stochastic error structure determined from replicated impedance measurements is independent of the type of measurement model used. While the regressed parameters may not be associated unequivocally with a set of deterministic or theoretical parameters for a given system, the measurement-model approach has been shown to represent adequately the impedance spectra obtained for a large variety of electrochemical systems. Regardless of their interpretation, the measurement-model representation can be used to filter and thus identify the nonstationary (drift) and high-frequency (noise) components contained in an impedance spectrum.

The measurement model has been applied in previous work to assess the error structure of a variety of systems including electrohydrodynamic impedance, electrochemical impedance data for reduction of ferricyanide on a Pt rotating disk, for corrosion of cast iron in Evian water, for corrosion of aluminum in orange juice, and for charging of electroactive polymers. The objective of the present work is to apply the error analysis approach to electrochemical impedance data collected for PEM fuel cells.

Experimental

Materials and chemicals.— The membrane electrode assembly (MEA) employed a 0.0308 mm (2 mil) thick Nafion N112 membrane. The catalyst layers of the MEA were platinum supported on carbon with a Pt catalyst loading of 0.4 mg/cm² on both the anode and the cathode sides. The material of the flow channel used was graphite with a single-channel horizontal serpentine flow configuration with the outlet lower than the inlet to facilitate removal of condensed water. Hydrogen gas was used as fuel and compressed air was used as oxidant. Compressed N₂ was used for purging of the fuel cell before and after experiments. A Barnstead E-Pure water system was used as a source of deionized water delivered to the anode and cathode humidifiers. The active surface area of the MEA was 5 cm².

A Scribner Associates 850C fuel cell test station was used to control reactant flow rates and temperatures. The test station was connected to a computer by an interface for data acquisition. The gas flow to the anode was held at a temperature of 40 ± 0.1°C, and the gas flow to the cathode was held at a temperature of 35 ± 0.1°C. The gas flows were humidified to 100% relative humidity at the respective temperatures. The cell temperature was held at 40 ± 0.1°C. The hydrogen flow rate was 0.1 L/min and the air-flow rate was 0.5 L/min.

Electrochemical impedance measurements.—Impedance measurements were performed using two different systems. The Scribner Associates 850C fuel cell test station contains both an electronic load and a frequency response analyzer. Impedance measurements obtained with the 850C were compared to impedance collected using a Gamry Instruments FC350 impedance analyzer coupled with a Dynaload electronic load RBL 100V-60A-400W. All electrochemical measurements were performed with a two-electrode cell in which the anode was used as a pseudo-reference electrode.

The protocol recommended by Ramani et al. was used to ensure that the system reached steady-state operation before impedance measurements were taken. The protocol consisted of two steps.

1. Upon startup, the current was swept from zero to the maximum value in forward and reverse directions until hysteresis in the polarization curve was no longer evident. This procedure was intended to ensure complete hydration of the MEA. This step required up to 48 h (break-in time) for a new MEA and 1.5 h for a system that had been recently used.

2. Once the hysteresis in the polarization curve was no longer evident, the current was set and the potential was monitored. Impedance measurements were conducted after the potential was stabilized. This step required 30 min.

The polarization curves were obtained by stepping the current from zero to the maximum current with an increment of 10 mA/30 s. A typical polarization curve is presented in Fig. 1.

Repeated impedance measurements were performed at several points on the polarization curve. The impedance measurements were conducted in galvanostatic mode for the frequency range of 3 kHz to 1 mHz with a 10 mA peak-to-peak sinusoidal perturbation. The corresponding potential perturbation ranged from 0.04 mV at high frequency to 0.4 mV at low frequency. The frequencies were spaced in logarithmic progression with 10 points per frequency de-
Cade. Impedance scans were conducted in autointegration mode with a minimum of 2 cycles per frequency measured. Each scan required 5 h for the Scribner system and 3 h for the Gamry system. The difference in time required can be attributed to differences in impedance settings.

The time required to make the measurement at each frequency is shown in Fig. 2. The long time required at lower frequencies made measurements at these frequencies susceptible to being influenced by nonstationary behavior.

Results

Representative impedance scans and error analysis are presented in this section. The stochastic error structure obtained from replicated measurements was used to weight subsequent regressions to assess consistency with the Kramers–Kronig relations. The experimentally determined stochastic error structure was also used to assess the quality of the regressions and to calculate the confidence interval for model predictions.

Electrochemical impedance response.— Impedance measurements were obtained at several points along the polarization curve presented, for example, in Fig. 1. Two different trends in the impedance response were observed. As shown in Fig. 3a, the impedance decreased with increasing current density for low current density ($i < 0.14 \, \text{A/cm}^2$). As shown in Fig. 3b, the impedance increased with increasing current density for high current density ($i > 0.14 \, \text{A/cm}^2$). These trends are consistent with changes in the slope of the polarization curve with current density.

The error analysis procedure described by Agarwal et al. was applied to sets of repeated impedance spectra. The procedure is illustrated in the subsequent sections for five repeated impedance spectra, shown in Fig. 4, collected at a current density of 0.2 A/cm$^2$ with the FC350.

Evaluation of stochastic errors.— Following the procedure described by Agarwal et al., the measurement model explained in Eq. 2 was fitted to each spectrum shown in Fig. 4 using a frequency-independent weighting. The number of parameters was constrained by the need to have the same number of parameters for each spectrum and the requirement that no parameter had a ±2σ (95.4%) confidence interval that included zero. Typically, 6 Voigt elements could be regressed to a spectrum. The standard deviation of the residual errors was used as an estimate for the standard deviation of the stochastic measurement errors. The same procedure was applied to impedance measurements collected using the Scribner 850C. The results are presented in Fig. 5a. Here, the comparison of results of the error analysis is based on the impedance data collected at 0.2 A/cm$^2$ for both the 850C and the FC350.

The level of stochastic errors was very similar for the impedance collected using the Gamry FC350 and the Scribner 850C. Standard deviations normalized by the modulus of the impedance are presented in Fig. 5b. As shown in Fig. 5b, the noise level of the measurements varied with frequency but was generally less than 0.3% of the modulus.

A similar procedure was applied to determine the structure of stochastic errors in impedance measurements collected at different currents along the polarization curve. For a given system, an error structure model could be determined following the general model described by Orazem et al., i.e.

![Figure 2. Average time required for impedance measurement at each frequency. The error bars associated with the standard deviation obtained from four experiments is smaller than the symbols used in the figure.](image)

![Figure 3. Impedance response obtained with the FC350: (a) low current density and (b) high current density.](image)

![Figure 4. Five scans of impedance data collected at a current density of 0.2 A/cm$^2$ with the FC350.](image)
where $R_m$ is the current measuring resistor corresponding to a given current range and $\alpha$, $\beta$, $\gamma$, and $\delta$ are constants determined for a given instrument and set of measurement parameters. For the Gamry FC350, all adjusted parameters were equal to zero with the exception of $\gamma = 0.679$. For the Scribner 850C, all adjusted parameters were equal to zero with the exception of $\alpha = 0.00213$ and $\gamma = 0.679$. Lines corresponding to Eq. 3 are given in Fig. 5a and b. Equation 3 was used to weight subsequent regressions to assess consistency with the Kramers–Kronig relations.

**Evaluation of high-frequency bias errors.**—In principle, a complex fit of the measurement model could be used to assess the consistency of impedance data. Sequential regression to either the real or the imaginary parts was shown to provide greater sensitivity to lack of consistency. The measurement model approach developed by Agarwal et al. was used to assess the consistency of high-frequency data with the Kramers–Kronig relations. The Voigt model was fitted to the real part of the measurement with a weighting based on the experimentally determined stochastic error structure. The parameters so obtained were then used to predict the imaginary part of the measurement, and a confidence interval for the prediction was calculated based on the estimated confidence intervals for the regressed parameters. Data that fell outside of the confidence interval were deemed to be inconsistent with the Kramers–Kronig relations.

This process is illustrated in Fig. 6 for the second impedance scan shown in Fig. 4. The fit to the real part of the impedance is given in Fig. 6a where the thin solid lines represent the confidence interval for the regression. The prediction of the imaginary part of the measurement is given in Fig. 6b. The prediction of the imaginary part of the impedance is excellent at intermediate frequencies, but a discrepancy is seen at high and low frequencies. Regression to the real part of the impedance generally provides fewer parameters than does regression to the imaginary part. For this reason, the discrepancy seen at low frequencies was not considered to be significant. The discrepancy at high frequency is seen where the real part of the impedance approaches asymptotically a finite value corresponding to a solution resistance.

The discrepancy is seen more clearly in the plots of normalized residual error given in Fig. 7a for the fitting errors and in Fig. 7b for the prediction errors. The normalization by the experimental value of the impedance causes the confidence-interval lines shown in Fig. 7b to tend toward $\pm \infty$ at the point where the imaginary impedance...
changes sign. The analysis shows that the nine highest frequencies fell outside the 95.4% confidence interval. These data were removed from the regression set. The conclusion that these points were inconsistent with the Kramers–Kronig relations is supported by the observation that the number of parameters that could be obtained from a complex regression increased when the high-frequency data were removed. In other words, deletion of data that were strongly influenced by bias errors increased the amount of information that could be extracted from the data. The bias in the complete data set induced correlation in the model parameters, which reduced the number of parameters which could be identified. Removal of the biased data resulted in a better conditioned data set that enables reliable identification of a larger set of parameters.5

A similar analysis was performed for the first and second measurements obtained by both the FC350 and 850C instruments. For all measurements, data measured at frequencies above 1000 Hz were found to be inconsistent with the Kramers–Kronig relations. These data were removed from the data used in subsequent regressions.

It is important to note that removal of data for which the imaginary impedance had a positive value was not sufficient to eliminate inconsistency with the Kramers–Kronig relations. As shown in Fig. 8a, the influence of the artifact extended well into the domain in which the imaginary impedance had a negative value. The filled symbols correspond to data that were deemed inconsistent with the Kramers–Kronig relations. The result may be seen as well in the Nyquist plot given as Fig. 8b.

**Evaluation of low-frequency bias errors.—** To test the consistency of the impedance data at low frequency, the imaginary part of

![Figure 7](image7.png)

**Figure 7.** Normalized residual errors for the regression presented in Fig. 6: (a) fit to the real part, where dashed lines represent the ±2σ bound for the stochastic error, and (b) prediction of the imaginary part, where solid lines represent the 95.4% confidence intervals for the model obtained by Monte Carlo simulations.

![Figure 8](image8.png)

**Figure 8.** Detailed representation of impedance data showing the inconsistency observed at high frequency: (a) expanded view of Fig. 6b and (b) expanded view of a Nyquist representation (see Fig. 4 for a complete spectrum). The filled symbols correspond to data that were deemed inconsistent with the Kramers–Kronig relations.
the impedance data was fitted using a weighting strategy based on the empirical model for error structure given as Eq. 3. The parameter set so identified was used to predict the real part of the impedance. The confidence interval for the prediction was obtained by a Monte Carlo simulation based on the confidence interval of the regressed parameters. The procedure is described by Agarwal et al.5

The Voigt measurement model was regressed to the imaginary part of the impedance data corresponding to the first scan of the impedance data presented in Fig. 4. The results, given in Fig. 9a, show that the measurement model could provide an excellent fit to the imaginary part of the data, even at the low frequencies that revealed inductive loops, characterized by positive values of imaginary impedance. The parameter values obtained from regression to the imaginary part of the impedance were used to predict the real part, as shown in Fig. 9b. The solid lines shown in Fig. 9b represent the upper and lower bounds of the 95.4% (2σ) confidence interval obtained for the model prediction. The low-frequency data that are outside the confidence interval can therefore be considered inconsistent with the Kramers–Kronig relations.

A more precise view of the regression quality and the level of agreement with the predicted values can be seen in plots of residual errors. The normalized residual error for the regression to the imaginary part of the impedance is shown in Fig. 10a, where the dashed lines indicate upper and lower bounds for the stochastic noise level for the measurement. The dashed lines were calculated as ±2σ, where σ was obtained from Eq. 3. The normalization by the experimental value of the impedance causes the dashed lines to tend toward ±∞ at the point where the imaginary impedance changes sign. The quality of the regression is indicated by the observation that the residual errors for the regression fall within the noise level of the experiment. The normalized residual errors for the predicted real value are shown in Fig. 10b, where the solid line represents the upper and lower bounds of the 95.4% (2σ) confidence interval obtained for the model prediction. A lack of agreement between predicted and experimental values is seen for frequencies below 0.05 Hz. The data for the four lowest frequencies are seen to fall outside the confidence interval for the prediction. These points can be described as being inconsistent with the Kramers–Kronig relations.

Similar bias-error analyses were performed for subsequent impedance scans. Figure 11a, for example, shows the normalized residual error for the imaginary part of the second scan, and Fig. 11b shows associated predicted error in the real part of the second scan. The agreement between predicted and experimental values is better for the second scan, shown in Fig. 11b, than for the first, shown in Fig. 10b. All data shown in Fig. 11b fall inside the 95.4% confidence interval.
The second and subsequent scans were found to be consistent with the Kramers–Kronig relations. The measurement model was also used to test the impedance data collected with the 850C for consistency with the Kramers–Kronig relations. In this case as well, some low-frequency data were found to be inconsistent with the Kramers–Kronig relations for the first of a series of repeated measurements. All data in the second and subsequent scans were found to be consistent with the Kramers–Kronig relations. The fit of the Voigt measurement model to the imaginary impedance data for the second scan of the series, for example, is shown in Fig. 12a. The predicted value for the real part of the impedance is compared to experimental values in Fig. 12b. The corresponding plots of normalized residual error are given in Fig. 13a and b. The data were found to be consistent with the Kramers–Kronig relations at all frequencies below 1000 Hz.

Impedance response after error analysis.— An example is presented in Fig. 14 of the results of a complex regression of the measurement model to a data set in which data were removed that were found to be inconsistent with the Kramers–Kronig relations. The measurement model was also used to test the impedance data collected with the 850C for consistency with the Kramers–Kronig relations. In this case as well, some low-frequency data were found to be inconsistent with the Kramers–Kronig relations for the first of a series of repeated measurements. All data in the second and subsequent scans were found to be consistent with the Kramers–Kronig relations. The fit of the Voigt measurement model to the imaginary impedance data for the second scan of the series, for example, is shown in Fig. 12a. The predicted value for the real part of the impedance is compared to experimental values in Fig. 12b. The corresponding plots of normalized residual error are given in Fig. 13a and b. The data were found to be consistent with the Kramers–Kronig relations at all frequencies below 1000 Hz.

Figure 11. Normalized residual errors for the fit of the measurement model to the second scan of impedance data presented in Fig. 4: (a) fit to the imaginary part, where dashed lines represent the ±2σ bound for the stochastic error, and (b) prediction of the real part, where solid lines represent the 95.4% confidence intervals for the model obtained by Monte Carlo simulations.

Figure 12. Regression of the Voigt model to the imaginary part of the impedance for the second scan of the impedance data collected at 0.2 A/cm² with the 850C: (a) fit to the imaginary part of the measurement and (b) prediction of the real part. The ○ represents experimental data, the thick solid lines represent the measurement model fit, and the thin solid lines represent confidence intervals.
measurement-system artifacts. As seen in Fig. 8, the influence of the artifact extended well into the domain in which the imaginary impedance had a negative value. Removal of data with a positive imaginary impedance is not sufficient to eliminate the influence of high-frequency instrument artifacts in impedance measurements.

The lack of consistency of low-frequency data with the Kramers–Kronig relations, seen for the first impedance measurement in Fig. 10b, is likely associated with start-up transients. As described in the Electrochemical Impedance Measurements section, impedance measurements were conducted after repeated current-voltage cycling to hydrate the membrane. The first impedance scan was then measured after the specified current had been set for 30 min and the corresponding potential was stabilized. The impedance results indicate that the fuel cell had not reached steady-state operation, due perhaps to changes to the humidification of the membrane and/or changes in consumption of reactants in the flow channels. The second scan of the impedance data, (see Fig. 11b) showed improved consistency of low-frequency data with the Kramers–Kronig relations. These data were collected approximately after 3 h of cell operation at the specified current.

The established start-up procedures, including stabilization of potential at a set current, were not sufficient to ensure the steady-state condition. Impedance spectroscopy is seen to be much more sensitive to the condition of the fuel cell. This work demonstrates the utility of the measurement-model error analysis for identifying steady-state operation.

Not all low-frequency inductive loops were free of artifacts caused by nonstationary behavior, but once a steady-state operation was established, the low-frequency inductive loops were found to be consistent with the Kramers–Kronig relations. The low-frequency inductive loops in PEM fuel cells can therefore be attributed to the response of physical processes occurring within the fuel cell.

**Conclusions**

The impedance data for a PEM fuel cell were analyzed using a Voigt measurement model. The inductive loops found at low frequencies were found to be consistent with the Kramers–Kronig relation once the fuel cell achieved steady-state operation. The formalism of the measurement model error analysis provides a means for determining whether a steady state has been achieved.

The present work confirms that the low-frequency inductive loops can be attributed to processes occurring in the fuel cell. Kramers–Kronig-consistent inductive loops were observed in the entire range (current density) of operation of the fuel cell. The results were independent of the impedance instrumentation used. The possible attribution of these low-frequency inductive loops to hydrogen peroxide intermediates and platinum deactivation is presented elsewhere.40 The information provided in this work can give a new motivation to use impedance spectroscopy to identify fundamental processes occurring within the fuel cell.

**Acknowledgments**

This work was supported by the NASA Glenn Research Center under grant NAG 3-2930 monitored by Dr. Timothy Smith with additional support from Gamry Instruments.

University of Florida assisted in meeting the publication costs of this article.

**References**


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In this article, the units used for the real and imaginary parts of the impedance presented in Fig. 3, 4, 6, 8, 9, 12, and 14 should be \( \Omega \) instead of \( \Omega \text{ cm}^2 \) and the standard deviations presented in Fig. 5 and 9 should also be \( \Omega \) instead of \( \Omega \text{ cm}^2 \). The area of the membrane electrode assembly used was 5 \( \text{cm}^2 \). The authors express their appreciation to Professor Eliezer Gileadi (Tel-Aviv University, Israel) and Professor Barry MacDougall (National Research Council, Canada) for alerting us to this error.