

NRC Publications Archive Archives des publications du CNRC

Accelerated lifetime testing for proton exchange membrane fuel cells using extremely high temperature and unusually high load Zhang, Jianlu; Song, Chaojie; Zhang, Jiujun

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. / La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version acceptée du manuscrit ou la version de l'éditeur.

For the publisher's version, please access the DOI link below./ Pour consulter la version de l'éditeur, utilisez le lien DOI ci-dessous.

Publisher's version / Version de l'éditeur:

https://doi.org/10.1115/1.4003977

Journal of Fuel Cell Science and Technology, 8, 5, pp. 051006-1-051006-5, 2011-06-16

NRC Publications Record / Notice d'Archives des publications de CNRC:

https://nrc-publications.canada.ca/eng/view/object/?id=acdcd522-6b48-44d1-b0ca-a9d63f9ab381 https://publications-cnrc.canada.ca/fra/voir/objet/?id=acdcd522-6b48-44d1-b0ca-a9d63f9ab381

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at https://nrc-publications.canada.ca/eng/copyright READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site https://publications-cnrc.canada.ca/fra/droits LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des guestions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.





Accelerated Lifetime Testing for Proton Exchange Membrane Fuel Cells Using Extremely High Temperature and Unusually High Load

In this paper, two testing protocols were developed in order to accelerate the lifetime testing of proton exchange membrane (PEM) fuel cells. The first protocol was to operate the fuel cell at extremely high temperatures, such as 300 °C, and the second was to operate the fuel cell at unusually high current densities, such as 2.0 A/cm². A PEM fuel cell assembled with a PBI membrane-based MEA was designed and constructed to validate the first testing protocol. After several hours of high temperature operation, the degraded MEA and catalyst layers were analyzed using SEM, XRD, and TEM. A fuel cell assembled with a Nafion 211 membrane-based MEA was employed to validate the second protocol. The results obtained at high temperature and at high load demonstrated that operating a PEM fuel cell under certain extremely high-stress conditions could be used as methods for accelerated lifetime testing. [DOI: 10.1115/1.4003977]

Keywords: proton exchange membrane (PEM) fuel cells, accelerated lifetime testing, degradation, carbon oxidation and corrosion, Pt catalyst agglomeration/isolation/ sintering

1 Introduction

PEM fuel cells are promising energy conversion devices for portable, stationary, and automotive power applications, due to their low/zero emissions, high efficiency, and high energy density. Unfortunately, two major technical challenges—high cost and unsatisfactory reliability/durability—have hampered the commercialization of PEM fuel cells. In efforts to discover breakthrough solutions, research studies worldwide have been focused on these two areas, resulting in significant recent progress in both cost reduction and reliability/durability improvement [1].

Regarding the latter challenge, lifetime testing of fuel cells under designed operating conditions seems to be the only effective way to determine failure modes and develop mitigation strategies [2–10]. For example, Cleghorn et al. [3] reported a three-year continuous lifetime test, in which a single cell was operated at 0.8 A/cm² for 26300 hours at 70 °C, ambient pressure, and 100% relative humidity (RH). Sishtla et al. [4] operated a single cell fed with a reformate fuel at 0.4 A/cm² for 5000 hours. Liu et al. [6] reported a lifetime test at 0.8 A/cm² for over 8000 hours; and Knights et al. [8] reported lifetime testing results for PEM fuel cell stacks run for up to 3000 hours. However, such lifetime testing hour-by-hour is extremely time-consuming as well as costly. In order to speed up the testing and diagnosis process, accelerated lifetime testing is necessary and associated methods have to be developed accordingly.

Accelerated testing subjects fuel cells to stressful conditions in order to accelerate them into failure mode [11]. The goal of these tests is to obtain durability and performance information about fuel cells and fuel cell components without conducting testing over a long period [12]. Unfortunately, at the current stage of technology there are still no standardized protocols for accelerated fuel cell testing. Different groups therefore use different methods and parameters according to their own application requirements. The United States Department of Energy (DOE) has suggested several methods for stress-testing the durability of key fuel cell components such as catalysts and membranes. For example, one of the protocols recommended by the DOE is to operate a fuel cell at open circuit voltage (OCV), 90 °C, and 30% RH to evaluate the chemical stability of the membrane [12]. But the variety of potential fuel cell applications means that the requirements for durability differ from one application to another. As a result, a number of accelerating methods have been developed for PEM fuel cell testing, and can be roughly classified into two types: (1) cycling the fuel cell load (cell voltage or current density), temperatures, relative humidities, and gas composition [12] between two different levels of parameter values, including on/off conditions (dynamic operation) [12], and (2) operating the fuel cell under stressful conditions such as OCV, high temperature, and low relative humidity [6,12].

Using the dynamic strategy, Escribano et al. [13] performed cell load cycling between zero current density and 1 A/cm² with both dry fuel and oxidant (cycle A), and between zero current density and 0.8 A/cm² with dry fuel and partially or fully humidified air (cycle B) [13]. Makharia et al. [14] evaluated catalysts by cycling a fuel cell between 0.7 and 0.9 V for 30,000 cycles at 80 °C and 100% RH.

Using the stress strategy, Endoh et al. [15] investigated the degradation of membrane electrolyte assemblies (MEAs) by operating a fuel cell at OCV and different RHs for ~160 hours. Shim et al. [16] reported conducting a similar procedure for over 100 hours at 76 °C and 100% RH. Ohma et al.[17] analyzed membrane degradation in a fuel cell held at OCV for 48 hours at 90 °C and 30% RH. Xing et al. [18] used an OCV test to measure membrane stability for 40 hours at 80 °C and 70% RH. Aarhaug et al. [19] ran an OCV test on a PEM fuel cell for over 1400 hours at 40 °C and 100% RH. Recently, Ralph et al. [20] reported accelerated testing of reinforced membranes by holding the MEA at OCV for

Jianlu Zhang Chaojie Song

Jiujun Zhang¹

e-mail: jiujun.zhang@nrc.gc.ca

Institute for Fuel Cell Innovation, National Research Council of Canada, 4250 Wesbrook Mall, Vancouver, BC, Canada V6T 1W5

Copyright © 2011 by ASME

¹Corresponding author.

Contributed by the Advanced Energy Systems Division of ASME for publication in the JOURNAL OF FUEL CELL SCIENCE AND TECHNOLOGY. Manuscript received September 4, 2009; final manuscript received February 7, 2011; published online June 16, 2011. Assoc. Editor: Jacob Brouwer.

up to 850 hours at 80 °C and 100% RH. Zhai et al., using a polybenzimidazole (PBI)-based MEA, conducted a lifetime test at OCV and high temperature (150 °C) for 520 hours [21]. Compared with normal operating conditions of 70–80 °C and 100% RH, high temperatures (>90°C) and low RH (<75%) are considered accelerating conditions.

Operating PEM fuel cells under extremely stressful conditions, such as very high temperatures or very high loads, may accelerate material degradation and thereby, further reduce the time required for lifetime testing. In our recent work on high temperature PEM fuel cells, we found that the fuel cell performance degraded much more quickly at extremely high temperatures (e.g., 300 °C) and high current densities (e.g., 2.0 A/cm²). These results suggested that such testing procedures could be used as protocols for accelerated testing. In this paper, we report on our use of these two testing protocols, presenting both experimental observation and data analysis.

2 Experimental

2.1 Extremely High Temperature Testing. The single fuel cell hardware used for 300 °C operation was designed and fabricated in-house. Details of the design have been reported elsewhere [21]. The MEA, purchased from PEMEAS Fuel Cell Technologies, contained a H_3PO_4 -doped polybenzimidazole (PBI) membrane, and had an active area of 2.5 cm² and a total Pt catalyst loading of 1.7 mg/cm². A Fideris 100 W test station was modified for high temperature operation up to 300 °C and used throughout the fuel cell testing. Dry H_2 and air were used, with flow rates controlled at 0.02 L/min and 0.05 L/min, respectively. Because performance degradation occurred so rapidly at this temperature, only non-steady-state polarization data was obtained, using a Solartron potentiostat 1287 to linearly scan the cell voltage at a rate of 10 mV/s.

2.2 High Load/Temperature Testing. For high current density operation, single cell hardware with an active MEA area of 50 cm² was purchased from ElectroChem, Inc. The MEA was fabricated by hot-pressing together an anode, a Nafion 211 membrane, and a cathode. The anode and cathode consisted of Pt/Ru catalyst (20% PtRu/C, E-TEK) and Pt catalyst (40% Pt/C, E-TEK), respectively, with a total Pt loading of 1.0 mg.cm⁻². The flow rates of H₂ and air were controlled at stoichiometric numbers of 1.2 and 3, respectively. The operating backpressure was 30 psig and the RH at both sides was 100%. Steady-state polarization data was collected using a Fideris fuel cell test station controlled by FC Power software. Before the polarization curves were recorded, the fuel cell was conditioned at 80 °C, 30 psig, and 100% RH with a load of 1.0 A/cm² for four hours. The cell temperature was then increased to 95 °C, and the fuel and air humidifier temperatures were increased accordingly to maintain 100% RH. After this, a polarization curve at the beginning of lifetime (BOL) was recorded. Then, the fuel cell was held at 2 A/cm² for lifetime testing, interrupted at various lifetime intervals to collect polarization curves.

2.3 Failure Analysis for MEAs. The MEAs were characterized before and after the stress testing using a scanning electron microscope (SEM), an X-ray diffractometer (XRD), a transmission electron microscope (TEM), and contact angle measurements. A JEOL JSM-6400 scanning electron microscope was used for SEM measurements and analysis. The XRD measurements were taken on a Bruker D8 X-ray diffractometer equipped with a graphite monochromator and a vertical goniometer, using Cu-K_{α} radiation. The TEM micrographs were recorded on a Hitachi H-7600 transmission electron microscope operated at 120 kV. The samples were embedded in LR white resin (hard formula) in gelatine capsules, and the resin was then polymerized in an oven at 60°C for 24 hours. Sections (60–120 nm thickness) were cut with a diamond knife, collected onto formvar-coated copper grids, and coated with carbon. Contact angle measurements were taken on an Artray FTA1000 contact angle measurement instrument.

3 Results and Discussion

3.1 High Temperature Operation. In our initial cell design, the fuel cell performance at 300 °C was low and degradation occurred very rapidly [22,23]. By optimizing the cell design and other conditions, we improved the cell performance by increasing the voltage and reducing the degradation rate. However, the latter was still high at such an elevated temperature. Figure 1 shows the polarization curves of a fuel cell assembly with a PBI-membranebased MEA, operated at 300 °C, 0% RH, and ambient backpressure. Two curves represent the polarizations before and after 24 hours of operation. Initially, a cell voltage of 0.57 V at 0.6 A/cm² could be obtained. However, within 24 hours of operation, the cell voltage had decreased significantly, from 0.655 V at 0.3 A/cm² to 0.22 V. In our previous reports [22,23], just two-three hours of operation at such a high temperature would have caused a similar level of degradation. The slower degradation rate resulted mainly from improvements in cell sealing and bipolar plate modification. In addition, it was found that MEA variation affects degradation rates; in other words, different batches of MEAs might yield different degradation rates, particularly when a fuel cell is operated at temperatures \sim 300 °C and above.

In order to identify the degradation mode, we disassembled the cell after this 24-hour operation and then carried out SEM analysis on the degraded MEA. The SEM results showed that the thickness of the membrane remained the same and no obvious membrane damage could be detected, indicating that membrane degradation did not occur over that short testing period. This result is in agreement with the literature [24,25], where it has been reported that PBI membranes have high thermal stability, with a glass transition temperature of 420 °C and good mechanical properties.

Figure 2 shows a typical XRD pattern for the cathode catalyst before and after 300 °C operation. First, the graphite (002) peak for the catalyst carbon support at 22° was significantly reduced after degradation, indicating that at such a high temperature, the carbon support had been oxidized according to chemical and electrochemical oxidation, Reactions (1) and (2), respectively

$$\begin{array}{c} C+O_2 \rightarrow CO_2 \uparrow & (\mbox{chemical reaction}) \end{array} (1) \\ C+2H_2O \rightarrow CO_2 \uparrow + 4H^+ + \ 4e^- \ (\mbox{electrochemical reaction}) \end{array}$$

(2)

According to chemical reaction kinetics, both reactions should have been accelerated by increasing the temperature. Therefore, at



Fig. 1 Polarizations of a fuel cell assembled with PBI membrane (H_3PO_4 -doped)- based MEA, as a function of current density at 300 °C, 0% RH, and ambient backpressure. MEA active area: 2.5 cm². H_2 flow rate: 0.02 L/min; air flow rate: 0.05 L/min. Cell voltage scan rate: 10 mV/s.

051006-2 / Vol. 8, OCTOBER 2011

Transactions of the ASME



Fig. 2 XRD pattern of the cathode catalyst before and after high temperature operation at 300 $^\circ$ C for 24 hours

300 °C, Reaction (1) should have converted the carbon support to CO_2 at the cathode; Reaction (2) should then have occurred mainly at the cathode, due to the high electrode potential (especially when the fuel cell was operated at OCV or low current densities). On the anode side, the electrode potential was maintained at about 0 V (versus standard hydrogen electrode) in a hydrogen environment, making it unfavorable for both reactions to occur. Note that CO_2 is the final product for Reactions (1) and (2). At medium temperatures or lower cathode potentials, carbon oxidation can produce surface species (groups) such as quinone on the carbon support. Such an event would change the carbon surface from hydrophobic to hydrophilic, causing a water management problem (water flooding) in the catalyst layer.

The electrochemical oxidation reaction is schematically expressed in Fig. 3.

Figure 2 also shows the change in morphology of the Pt catalyst after operation at 300 °C. The diffraction peak became much sharper after high temperature operation, indicating an increase in Pt particle size. The Pt crystal size can be calculated by the following Eq. (3)

$$D = \frac{0.9\lambda}{\beta\cos\theta} \tag{3}$$

where *D* is the crystal size, λ is the wavelength, β is the full width at half-maximum (FWHM) of the diffraction peak, and θ is the Bragg diffraction angle. Before high temperature operation, the particle size was 3.6 nm. After 24 hours of 300 °C testing, the particle size increased to 6.0 nm. Pt particle agglomeration was also observed in the TEM image (Fig. 4). After 24 hours of operation at 300 °C, more large particles were found [Fig. 4(*b*) than had initially been present before testing [Fig. 4(*a*)].

It can also be observed from Fig. 4(b) that because the carbon support was gone, some of the Pt catalyst particles became iso-



Fig. 3 Schematic representation of carbon support degradation

Journal of Fuel Cell Science and Technology



Fig. 4 TEM photos of catalyst (a) before and (b) after 300 $^{\circ}$ C operation for 24 hours

lated from each other. This change could result in poor electrical connectivity and corresponding high resistance in the catalyst layer, an observation consistent with the literature [2–5, 7, 8, 10, 13–19, 21]. It has been reported that after lifetime testing and accelerated testing, the Pt particle size can increase significantly.

Carbon support oxidation, Pt size enlargement (also known as the Pt sintering effect), and Pt catalyst particle isolation, after only 24 hours of fuel cell operation, are consistent with observations reported after a long period of lifetime testing, indicating that 300 °C operation can be used as an effective accelerated testing protocol for rapid screening of fuel cell materials, particularly catalysts and catalyst supports.

3.2 High Current Density/Temperature Operation. Figure 5 shows the polarization curves of the fuel cell after operation at a high current density (2.0 A/cm²) at 95 °C, 30 psig backpressure, and 100% RH. It can be seen that as the operating time increased, the cell performance degraded significantly. The open circuit voltage (OCV), at the beginning of lifetime, was decreased from 0.95 V to 0.85 V after 94 hours of operation at high current density. The performance at low current density was also decreased. For example, at 0.4 A/cm², the cell voltage dropped from 0.77 V at BOL to 0.70 V after 94 hours of operation. The performance between the initial and after 94 hours of lifetime testing was decreased in a parallel way as in the low current density region $(< 0.4 \text{ mA/cm}^2)$. It is believed that this voltage drop at a low current density is partially induced by high temperature operation, causing chemical and electrochemical oxidation as described previously. At 95 °C, both carbon oxidation and Pt agglomeration and isolation could occur, decreasing the number of effective reaction sites and thereby, causing a drop in fuel cell performance. This may cause a decrease in the electroactive surface area, resulting in an OCV drop and a performance decrease as well in a parallel way. On the other hand, carbon oxidation in the electrode might produce surface groups, changing the surface from hydrophobic to hydrophilic and leading to inefficient water management. This is indicated by the change in the polarization curve at high current densities.

As shown in Fig. 5, in the high current density range, the performance dropped even more obviously after lifetime testing. For example, at a current density of 1.6 A/cm², the cell voltage was 0.623 V at the beginning of the test, then decreased to 0.592 V, 0.518 V, and 0.44 V after 12 hours, 24 hours, and 94 hours of testing, respectively. The mass transfer limit region shifted to a lower current density as the operation time increased. This shift might be attributable to water flooding resulting from changes in the electrode properties, caused by high current density and high temperature operation.

The voltage change over time during high current density operation is shown in Fig. 6. Large spikes (> 300 mV) can be observed after 50 hours of high load and high temperature operation, and these spikes occur more frequently as the operation time increases. After about 85 hours, water flooding became too significant to

OCTOBER 2011, Vol. 8 / 051006-3



Fig. 5 Polarization curves of a fuel cell after operation at high current density (2.0 A/cm²) at 95° C, 30 psig backpressure, and 100% RH

continue the testing. For comparison, an MEA operated at a lower current density of 1 A/cm² is also shown in Fig. 6. No large spikes can be observed before 90 hours, and the amplitude of the spikes is much smaller (< 100 mV) than at 2.0 A/cm² operation.

In order to confirm the spikes were caused by an increase in the electrode hydrophilicity, the contact angles of the electrode were measured before and after lifetime testing. Table I summarizes the contact angle results of the cathode electrode (0.7 mg/cm² Pt loading, 0.21 mg/cm² Nafion loading, with a back layer of 1 mg/cm² carbon coated on 10% PTFE wet-proof carbon paper) before and after high load (2 A/cm²) or low load (1 A/cm²) operation. At high load operation, the contact angle decreased significantly with increasing operating time. For example, after 94 hours of operation at 2 A/cm², the contact angle was reduced from 145.1 to 93.1degrees, and continued operation to 190 hours resulted in a further decrease to 75.7 degrees. For the MEA operated at 1 A/cm^2 , 120 hours of operation decreased the contact angle from 145.1 to 120.5 degrees. This suggests that the electrode became more hydrophilic after operation at 2 A/cm², resulting in severe water flooding and consequent cell performance degradation.

We believe that change in hydrophobicity may be mainly caused primarily by the back layer and the carbon fiber paper. In general, the catalyst layer is hydrophilic, and the back layer, as well as the carbon fiber paper, are hydrophobic in order to facilitate gas transport. It was reported that the back layer (or microporous layer) and carbon fiber paper could be oxidized and become more hydrophilic during lifetime testing and/or accelerated testing, leading to fuel cell performance degradation [26].

High load combined with high temperature operation can effectively speed up the degradation process of a fuel cell. It is believed that operating a high temperature fuel cell at a high current density simultaneously could cause an even higher local temperature, resulting in accelerated chemical or electrochemical oxidation of carbon both in the back layer and in the carbon paper, changing them from hydrophobic to hydrophilic. Such a change in the hydrophobicity of the electrode can cause water management problems (water flooding) and thus slow down the mass transfer process, resulting in fuel cell degradation.

The testing time in Fig. 6 is less than 100 hours, which is much shorter than a typical fuel cell lifetime test (from several thousand to more than ten thousand hours) under normal operating conditions. Therefore, high current density operation can be considered an accelerated testing method.

The comparison between the lengths of lifetime testing at the accelerated condition and at normal operation condition is of importance in validating the acceleration methods and quickly obtaining useful information about fuel cells lifetime behavior. Since a fair comparison with literature data is difficult because of different MEAs and operation conditions used, instead we did a rough comparison between our results and the data in literature [2]. At 1.0 A/cm², Cleghorn et al. [2] reported a cell voltage



Fig. 6 Voltage versus time plots at 95 $^\circ\text{C}$, 30 psig backpressure, and 100% RH, at current densities of 2.0 A/cm² and 1.0A/cm², respectively

decrease of 70 mV and 150 mV, respectively, after 10100, and 15000 hours of lifetime testing at 70°C. In this work, a cell voltage decrease of 100 mV was observed at 95°C. The 2.0 A/cm² and 95°C used in this work are more severe than the 0.8 A/cm² and 70°C used by Cleghorn at al. [2], indicating that the conditions used in this work are an accelerated testing protocol.

We also investigated the correlation between the testing accelerated by high load and that at normal load operation conditions. Figure 7 shows the steady-state polarization curve obtained after 24 hours of high load lifetime testing at 2.0 A/cm², and that after 130 hour at normal operation testing at 1.0 A/cm². At current densities less than 1.1 A/cm², the two curves after lifetime testing at two different current densities are almost identical. However, the curves start to separate at a current density 1.0 A/cm². The lifetime testing at a high current density (2.0 A/cm²) gives much lower cell voltages when compared to that obtained at normal conditions (1.0 A/cm²) even its lifetime test was only carried out for 24 hours. This result further indicates that the fuel cell degradation can be accelerated at higher operation loads. The testing at a current density of 2.0 A/cm² could give a five-times faster acceleration than that tested at 1.0 A/cm². It was confirmed by contact angle tests that this difference is due to the hydrophobicity change after high load operation as discussed in the previous section, which leads to a deteriorated mass transfer at high current densities.

4 Conclusions

Two testing protocols were developed in order to accelerate the lifetime testing of PEM fuel cells. The first protocol was to operate the fuel cell at an extremely high temperature such as 300 °C, and the other was to operate the fuel cell at an unusually high current density such as 2.0 A/cm². A fuel cell assembled with a PBI membrane-based MEA was designed and constructed for validating the first protocol, and the testing results obtained at 300 °C showed rapid fuel cell degradation. After 24 hours of high

 Table 1
 Contact angles of the MEA cathode catalyst layer (catalyst layer composition: Pt loading, 0.7 mg/cm²; Nafion loading, 0.21 mg/cm²)

Testing conditions	Contact angle (degrees)
Before testing	145.1
After 120 h running at 1.0 A/cm ²	120.5
After 94 h running at 2.0 A/cm ²	93.2
After 190 h running at 2.0 A/cm ²	75.7

051006-4 / Vol. 8, OCTOBER 2011

Transactions of the ASME



Fig. 7 Polarization curves of a fuel cell after operation at normal condition (1 A/cm²) for 130 hours and after operation at high current density (2.0 A/cm²) for 24 hours at 95° C, 30 psig backpressure, and 100% RH

temperature operation, the degraded MEA and catalyst layers were analyzed using SEM, XRD, and TEM, revealing that carbon oxidation and Pt agglomeration might be the main cause, suggesting that operating a PEM fuel cell under extremely high stress conditions such as 300 °C can be used as a method of accelerated lifetime testing. To test the second protocol of operating a PEM fuel cell at an unusually high load, a fuel cell assembled with a Nafion 211 membrane-based MEA was employed. Rapid degradation was also observed within 100 hours of testing at 2.0 A/cm² and 95 °C. Change in hydrophobicity in the back layer and carbon fiber paper was found to be the main reason. This high load protocol is thus also believed to be an accelerated testing method for PEM fuel cell lifetime testing.

Acknowledgment

This work was financially supported by the National Research Council of Canada (NRC) National Fuel Cell Program, and the NRC Institute for Fuel Cell Innovation. The authors also thank Dr. Ken Shi for the TEM, as well as Mr. Tom Vanderhoek and Mr. Andrew Mattie for hardware support.

References

- Garland, N. L., "Fuel Cells Sub-Program Overview," http://www.hydrogen. energy.gov/pdfs/progress08/v_0_fuel_cells_overview.pdf.
- [2] Cleghorn, S. J. C., Mayfield, D. K., Moore, D. A., Moore, J. C., Rusch, G., Sherman, T. W., Sisofo, N. T., and Beuscher, U., 2006, "A Polymer Electrolyte Fuel Cell Life Test: 3 Years of Continuous Operation," J. Power Sources, 158, pp. 446–454.
- [3] Wilson, M. S., Valerio, J. A., and Gottesfeld, S., 1995, "Low Platinum Loading Electrodes for Polymer Electrolyte Fuel Cells Fabricated Using Thermoplastic Ionomers," Electrochim. Acta, 40, pp. 355–363.

- [4] Sishtla, C., Koncar, G., Platon, R., Gamburzev, S., Appleby, A. J., and Velev, O., 1998, "Performance and Endurance of a PEMFC Operated with Synthetic Reformate Gas Feed," J. Power Sources, 71, pp. 249–255.
- [5] Isono, T., Suzuki, S., Kaneko, M., Akiyama, Y., and Yonezu, I., 2000, "Development of High Performance PEFC Module Operated by Reformate Gas," J. Power Sources, 86, pp. 269–273.
 [6] Liu, W., Ruth, K., and Rusch, G., 2001, "Membrane Durability in PEM Fuel
- [6] Liu, W., Ruth, K., and Rusch, G., 2001, "Membrane Durability in PEM Fuel Cells," J. New Mater. Electrochem. Syst., 4, pp. 227–231.
 [7] Xie, J., Wood, D. L., III, Wayne, D. M., Zawodzinski, T. A., Atanassov, P.,
- Xie, J., Wood, D. L., III, Wayne, D. M., Zawodzinski, T. A., Atanassov, P., and Borup, R. L., 2005, "Durability of PEFCs at High Humidity Conditions," J. Electrochem. Soc., 152, pp. A104–A113.
 [8] Knight, S. D., Colbow, K. M., St-Pierre, J., and Wilkinson, D. P., 2004,
- [8] Knight, S. D., Colbow, K. M., St-Pierre, J., and Wilkinson, D. P., 2004, "Aging Mechanism and Lifetime of PEFC and DMFC," J. Power Sources, 127, pp. 127–134.
- [9] Endoh, E., Terazono, S., and Widjaja, H., 2002, Abstract 89, The Electrochemical Society Meeting Abstracts, Vol. 2002-2, Salt Lake City, Utah, Oct 20-24.
- [10] Tada, T., Toshima, N., Yamamoto, Y., and Inoue, M., 2007, "Investigation of Catalyst Degradation After Single Cell Life Tests," Electrochemistry (Toyko, Jpn.), 75, pp. 221–230.
- [11] Pierpont, D., Hicks, M., Watschke, T., and Turner, P., 2006, "Accelerated Testing and Lifetime Modeling for the Development of Durable Fuel Cell MEAs," ECS Trans., 1, pp. 229–237.
- [12] Garland, N. L., Benjamin, T. G., and Kopasz, J. P., 2007, "DOE Fuel Cell Program: Durability Technical Targets and Testing Protocols," ECS Trans., 11, pp. 923–931.
- [13] Escribano, S., Morin, A., Solan, S., Sommacal, B., Capron, P., Rougeaux, I., and Gebel, G., 2005, "Study of MEA Degradation in Operating PEM Fuel Cells," 3rd European PEFC Forum, Lucern, Switzerland, July 4–8.
- [14] Makharia, R., Kocha, S. S., Yu, P. T., Sweikart, M. A., Gu, W., Wagner, F. T., and Gasteiger, H. A., 2006, "Durable PEM Fuel Cell Electrode Materials: Requirements and Benchmarking Methodologies," ECS Trans., 1, pp. 3–18.
- [15] Endoh, E., Terazono, S., Widjaja, H., and Takimoto, Y., 2004, "Degradation Study of MEA for PEMFCs under Low Humidity Conditions," Electrochem. Solid-State Lett., 7, pp. A209–A211.
- [16] Shim, J. Y., Tsushima, S., and Hirai, S., 2007, "Preferential Thinning Behaviours of the Anode Side of the PEM Under Durability Test," ECS Trans., 11, pp. 1151–1156.
- [17] Ohma, A., Yamamoto, S., and Shinohara, K., 2007, "Analysis of Membrane Degradation Behaviour During OCV Hold Test," ECS Trans., 11, pp. 1181–1192.
 [18] Xing, D., Zhang, H., Wang, L., Zhai, Y., and Yi, B., 2007, "Investigation of
- [18] Xing, D., Zhang, H., Wang, L., Zhai, Y., and Yi, B., 2007, "Investigation of the Ag-SiO₂/Sulfonated Poly(Biphenyl Ether Sulfone) Composite Membranes for Fuel Cell," J. Membr. Sci., 296, pp. 9–14.
- [19] Aarhaug, T. A., and Svensson, A. M., 2006, "Degradation Rates of PEM Fuel Cells Running at Open Circuit Voltage," ECS Trans., 3, pp. 775–780.
- [20] Ralph, T. R., Barnwell, D. E., Bouwman, P. J., Hodgkinson, A. J., Petch, M. I., and Pollington, M., 2008, "Reinforced Membrane Durability in Proton Exchange Membrane Fuel Cell Stacks for Automotive Applications," J. Electrochem. Soc., 155, pp. B411–B422.
- [21] Zhai, Y., Zhang, H., Xing, D., and Shao, Z., 2007, "The Stability of Pt/C Catalyst in H₃PO₄/PBI PEMFC During High Temperature Life Test," J. Power Sources, 164, pp. 126–133.
- [22] Tang, Y., Zhang, J., Song, C., and Zhang, J., 2007, "Single PEM Fuel Cell Design and Validation for High Temperature MEA Testing and Diagnosis up to 300°C," Electrochem. Solid-State Lett., 10, pp. B142–B146.
- [23] Song, C., 'Electrochem. Solid-State Lett., 10, pp. B142–B146.
 [23] Song, C., Hui, R., and Zhang, J., 2007, 'High Temperature PEM Fuel Cell Catalysts and Catalyst Layers,' *PEM Fuel Cell Electrocatalysts and Catalyst Layers*, J. Zhang, ed., Springer-Verlag, London, Chap. 18.
- [24] Cheng, C., Luo, J., Chuang, K. T., and Sanger, A. R., 2005, "Propane Fuel Cells Using Phosphoric-Acid-Doped Polybenzimidazole Membranes" J. Phys. Chem. B, 109, pp. 13036–13042.
- [25] Musto, P., Karasz, F. E., and MacKnight, W. J., 1993, "Fourier Transform Infra-Red Spectroscopy on the Thermo-Oxidative Degradation of Polybenzimidazole and of a Polybenzimidazole/Polyetherimide Blend," Polymer, 34, pp. 2934–2945.
- [26] Wood, D. L., III and Borup, R. L., 2009, "Durability of Gas-Diffusion and Microporous Layers," *Polymer Electrolyte Fuel Cell Durability*, Buchi, F. N., Inaba, M., and Schmidt, T. J., eds., Springer, New York.