



A Comparison of Bis[(perfluoroalkyl)sulfonyl]imide Ionomers and Perfluorosulfonic Acid Ionomers for Applications in PEM Fuel-Cell Technology

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Two structurally related perfluorinated ionomer materials, one a conventional sulfonic-acid-based ionomer (Nafion) and the other an experimental bis[(perfluoroalkyl)sulfonyl]imide-based ionomer in which the sulfonic acid group has been replaced by a sulfonyl imide acid group, were studied in parallel to evaluate their relative utility as membrane materials for use in polymer electrolyte membrane (PEM) fuel cells. Studies focused on membrane ionic conductivity and water content under varying conditions of relative humidity, and on device-level fuel-cell tests using membrane-electrode assemblies (MEAs) fabricated from membranes of the two ionomers. The overall finding is that the two ionomer materials behave similarly with respect to their electrochemical properties and performance in PEM fuel-cell devices. In one instance, a sulfonyl-imide-based MEA exhibited substantially improved performance relative to a comparable Nafion-based MEA in fuel-cell tests. The improvement is probably attributable to a combination of favorable materials properties and membrane thickness effects.

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Perfluorinated ionomers such as DuPont's Nafion and other closely related perfluorosulfonic acid (PFSA) ionomers are leading candidates for use as membrane materials in polymer electrolyte membrane (PEM) fuel cells. They possess many of the desirable qualities required for a successful PEM fuel cell, namely, high protonic conductivity, good mechanical properties, and excellent long-term chemical stability. Despite the desirable qualities of these materials, however, they also possess certain limitations, among which is a tendency toward diminished protonic conductivity under conditions of low water availability.¹

Several members of a related class of ionomers based on the bis[(perfluoroalkyl)sulfonyl]imide acid group have been synthesized and characterized by DesMarteau and co-workers.²⁻⁴ The perfluorosulfonyl imide group is known to possess stronger gas-phase acidity⁵ and improved thermal stability relative to the perfluorosulfonic acid moiety. Additionally, enhanced oxygen reduction kinetics have been reported in phosphoric acid fuel cells (PAFCs) when a small amount of a monomeric bis[(perfluoroalkyl)sulfonyl]imide species was used as an electrolyte additive.⁶ Recent work by Sumner and co-workers has established that sulfonyl imide-based ionomers (Fig. 1b) exhibit ionic conductivities that are at least as high as that of Nafion (Fig. 1a) ionomers of similar structure and equivalent weight.^{4,7} Thus, there is reasonable expectation that ionomer materials based on the perfluorosulfonyl imide moiety are good candidates for use as membranes in PEM fuel cells.

Work presented in this paper explores the similarities and differences between perfluorosulfonylimide ionomers and perfluorosulfonic acid ionomers as membrane materials for use in PEM fuel cells. Membrane-electrode assemblies (MEAs) were fabricated from Nafion and the experimental sulfonylimide ionomer membranes using the thin-film decal transfer method.⁸ Performance of the MEAs was tested in a single-cell fuel-cell test fixture. To place these fuel-cell tests in context, the ionomers were also characterized with respect to their water content and ionic conductivity as a function of relative humidity.

Experimental

Ionomer synthesis and membrane preparation.—Nafion 117 and 1135 membranes were obtained commercially (Solution Technology, Inc., Mendenhall, PA) and were cleaned using a multistep procedure involving boiling in a 3% aqueous hydrogen peroxide solution fol-

lowed by boiling in deionized water for 1 h. The sulfonyl imide ionomer materials were prepared by copolymerization of tetrafluoroethylene and a perfluorinated vinyl ether monomer (as the sodium salt) using a continuous addition semibatch redox-initiated aqueous emulsion polymerization technique.² Membranes were prepared by solution casting of the ionomer in the tetraalkylammonium salt form from a water/methanol/*N,N*-dimethylformamide solvent mixture. The solvents were removed at 80°C under 75 kPa vacuum followed by annealing at 220–250°C for 4 h. The membranes were cleaned and converted to the proton form via sequential treatment with hot concentrated nitric acid, deionized (DI) water, 3% hydrogen peroxide, hydrochloric acid, and again DI water.

Ionic conductivity and water uptake measurements.—Ionomer conductivities were measured at ambient temperature (25 ± 1°C) as a function of the relative humidity of the environment using a two-electrode cell similar to that described by Zawodzinski and co-workers.⁹ The cell had a 1 cm path length and utilized four platinumized Pt foil electrodes (two on each side) poised to make electrical contact to the two ends of an ionomer strip approximately 3 cm long and 1 cm wide. Ionomers were pretreated by boiling in 70% nitric acid followed by boiling in deionized water, after which they were mounted in the conductivity cell. The membrane/cell assembly was suspended over saturated salt solutions in sealed chambers to provide for control of the relative humidity of the environment.^{10,11} DI water, saturated (NH₄)₂SO₄, NaBr, CaCl₂, and KOH solutions, and solid P₄O₁₀ were used to produce 100, 81, 58, 31, 9, and 0% relative humidity environments, respectively. Membrane impedance was measured between 100 and 20,000 Hz using an electrochemical analyzer (model 1280B, Solartron, Allentown, PA) controlled with ZPlot for Windows software (version 2.1a, Scribner, Southern Pines, NC). Measurements were made in potentiostatic mode with the dc bias set to zero and the ac amplitude set to between 1 and 100 mV. (The higher amplitudes were used for low-conductivity samples for which lower amplitudes did not produce signals large enough to be detected.) Conductivities were calculated from the measured impedance in the frequency-independent region (usually between 1000 and 10,000 Hz where the effects of contact impedance are minimal) using the cell path length, the membrane width, and the membrane thickness obtained using a micrometer.

The water content of ionomer samples exposed to different relative humidity environments was measured gravimetrically with a conventional analytical balance at room temperature. Ionomer samples were first soaked in DI water for 2 h and weighed. The samples were then suspended in a specific relative humidity environment in a closed vessel for 90 min after which they were re-

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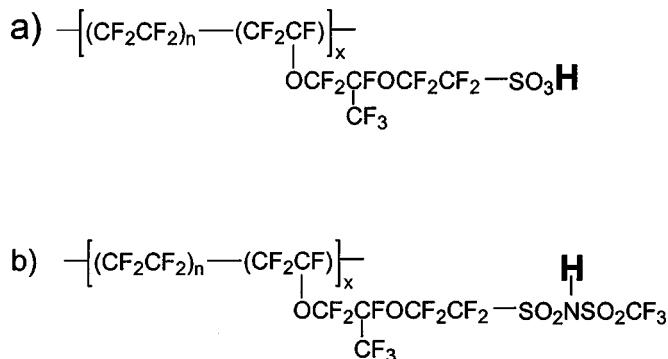


Figure 1. Primary structure of perfluorinated ionomers: (a) DuPont Nafion and (b) bis[perfluoroalkyl]sulfonimidic acid.

moved and quickly weighed. Exposure times longer than 90 min did not result in appreciable changes in mass. The measured masses were found to be stable over the few minutes that it took to obtain a measurement, which suggests that the mass is indicative of the water content of the ionomer in the controlled humidity environment and not that of the environment to which the samples were briefly exposed while they were being weighed. This process was repeated for each of a series of different humidity environments, proceeding from high to low humidity. The masses of water-free ionomers were obtained by drying the samples at 110°C under vacuum overnight followed by weighing and were used to calculate water contents under humid conditions by difference.

MEA preparation and testing.—Five membrane electrode assemblies were prepared using the decal transfer method developed by Wilson and co-workers at Los Alamos National Laboratory.⁸ An ink containing Pt/C and ionomer was prepared by mixing catalyst (20% Pt on Vulcan XC-72R, Alfa Aesar, Ward Hill, MA) with an appropriate amount of solubilized Nafion ionomer (1100 EW, Solution Technology, Mendenhall, PA) in a small vial with a magnetic stirrer. The dry mass ratio of supported catalyst to ionomer was 5:2. After a few hours of stirring, the proton form of the ionomer was converted to the tetrabutyl ammonium form (TBA^+) with the addition of TBAOH (1 M in methanol, Aldrich). The mixture was thickened by addition of glycerol. Additional changes in consistency were made by adding isopropanol or by allowing the evaporation of lighter alcohols. Once the desired consistency was achieved, the ink was applied by painting in thin layers onto Teflon-coated templates to which a dry release agent (6075 dry film lubricant, Crown, Woodstock, IL) had been applied. Catalyst loadings in the range of 0.3 mg Pt/cm² were used for both the anode and cathode of these MEAs by a process of sequential ink painting/drying steps.

Benchmark MEAs were fabricated using commercially available Nafion 117 or 1135 membranes (175 and 88 µm thick, respectively, both of equivalent weight 1100 g equiv⁻¹). Three test MEAs were fabricated using the experimental sulfonylimide ionomer membranes. One membrane was fabricated from a sulfonylimide ionomer having an equivalent weight of 1200 g equiv⁻¹ (as determined by titration of the ionomer prior to casting as a membrane) and a

thickness of 150 µm. The other two imide membranes were from the same polymerization batch and therefore shared an equivalent weight of 1075 g equiv⁻¹ and thickness of 60 µm. All membranes were pretreated prior to use in fabricating MEAs by boiling in 0.5 M NaOH to convert from the proton form to the sodium form.

During fabrication of all MEAs, the membrane was first pulled dry and flat on a vacuum table at 120°C for 15 min. The painted catalyst templates were then hot-pressed onto the membrane at 200–210°C and 600 lb pressure for 5 min. Negligible catalyst remained on the templates when the pressing was complete and the Teflon-coated templates were peeled away. Following pressing, the MEA was boiled in 0.5 M sulfuric acid for 30 min to convert all ionomer back to the proton form. The acid solution was allowed to cool to room temperature, with the MEA left soaking in the solution overnight. Following acid treatment, the MEA was washed several times with deionized water and then boiled for 15 min in deionized water. The MEA was then pulled dry and flat on a vacuum table at 60°C for 20 min prior to being mounted in the fuel-cell hardware.

The MEA was sandwiched between the two graphite blocks and fiberglass-based gaskets. On each side of the MEA was placed a piece of uncatalyzed gas diffusion backing (ELAT/NC, E-TEK, Natick, MA). The fixture was heated using cartridge heaters. Heated humidified fuel (H₂) and oxidant (O₂) gases were supplied to the test fixture by the test station. Typical cell temperatures were 80–110°C. Water-filled gas sparger bottles were set to temperatures 10–20°C higher than the cell temperature to deliver well-humidified gases to the cell. The gas flow rates were based on the current generated by the cell; gas flows in the range of four to five times stoichiometric were used. The cell was operated with 2 atm of back pressure on both the anode and cathode.

Cells were broken in at a cell voltage of 0.5 V overnight, after which a series of polarization curves was acquired in controlled voltage mode under different operating conditions (cell temperature, gas humidification). The cell open circuit voltage (OCV) was close to or above 1.0 V for all MEAs. The cell resistance was monitored during acquisition of the polarization curves using the current-interrupt method.

Crystallinity measurements.—Powder X-ray diffraction (XRD) measurements were made using a Scintag XDS/2000 0-0 diffractometer with CuK α_1 radiation ($\lambda = 1.5406 \text{ \AA}$) and an intrinsic germanium solid-state detection system. Step scans (at 1 s/step with a step size of 0.01°/step) were taken between 10 and 24° (20) on 5 × 5 mm membrane samples on a zero-background quartz sample pan.

Results and Discussion

Ionomer conductivity and water content.—Table I presents data on the ionic (proton) conductivity and water content at selected values of relative humidity for Nafion membranes (1100 g equiv⁻¹) and two imide membranes (1200 and 1075 g equiv⁻¹). As expected, water content and ionic conductivity decrease precipitously with decreasing relative humidity for all of the materials. The data for Nafion are in good quantitative agreement with previously reported values from several laboratories,^{12–14} which serves to validate the

Table I. Water uptake and conductivity for Nafion 1100, Imide 1075, and Imide 1200.

Relative humidity (%)	H ₂ O	Conductivity (S/cm)	H ₂ O	Conductivity (S/cm)	H ₂ O	Conductivity (S/cm)
100	19	6.18×10^{-2}	49	5.2×10^{-2}	28	5.3×10^{-2}
81	10	2.06×10^{-2}	22	2.1×10^{-2}	8	8.5×10^{-3}
58	5	8.03×10^{-3}	3	5.1×10^{-3}	5	2.7×10^{-3}
31	3	1.38×10^{-3}	2	4.6×10^{-4}	4	3.8×10^{-4}
9	1	1.12×10^{-4}	1	6.2×10^{-5}	1	5.9×10^{-5}

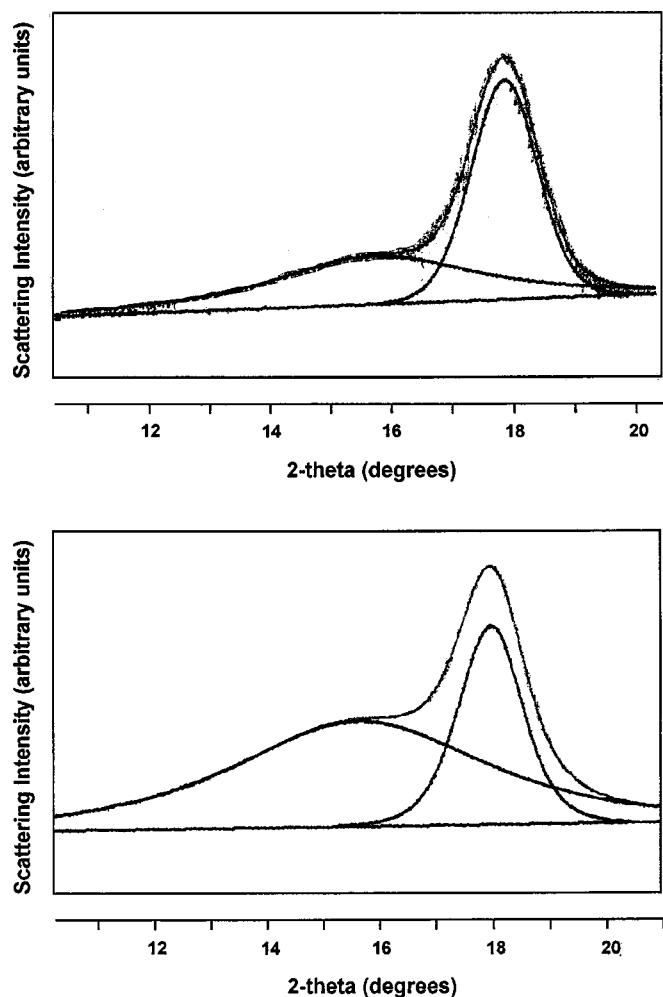


Figure 2. Powder XRD data for sulfonyl imide ionomers: (top) sulfonyl imide EW 1075 and (bottom) 1200.

measurement technique. The water content and conductivity for the imide ionomers are slightly lower than values that have previously been reported for nominally similar materials.⁴ This variation is thought to reflect the fact that the experimental protocols used for production of the imide ionomers are continually evolving, and therefore they yield materials with slightly different properties. Even so, the general trends of decreasing water content and conductivity with decreasing humidity are quite similar in Nafion and all of the imide ionomers tested. Such behavior in Nafion has been attributed to the effects of ion clusters and channels for which the size and connectivity are diminished as humidity and water content decrease.¹⁵ It is not unexpected that similar behavior would be found in the imide and Nafion ionomers, because the two ionomers differ only in the chemical nature of the acid group.

It appears that the differences in water absorption and conductivity exhibited by the two sulfonyl imide ionomers are correlated with internal structural factors as demonstrated by the powder XRD data presented in Fig. 2. The imide 1200 sample was found to have a crystallinity of 54%, almost double the 30% PTFE crystallinity seen for the imide 1075 sample. The greater degree of PTFE crystallinity for the 1200 EW sample would hamper the ionomer's ability to expand, which in turn would negatively affect both water absorption and ionic conductivity.

Fuel cell testing.—Given the similarities in structure and properties between Nafion and the imide ionomers, it is of interest to compare the behavior of the two materials for applications as membranes in PEM fuel-cell technology. Based on thickness and equiva-

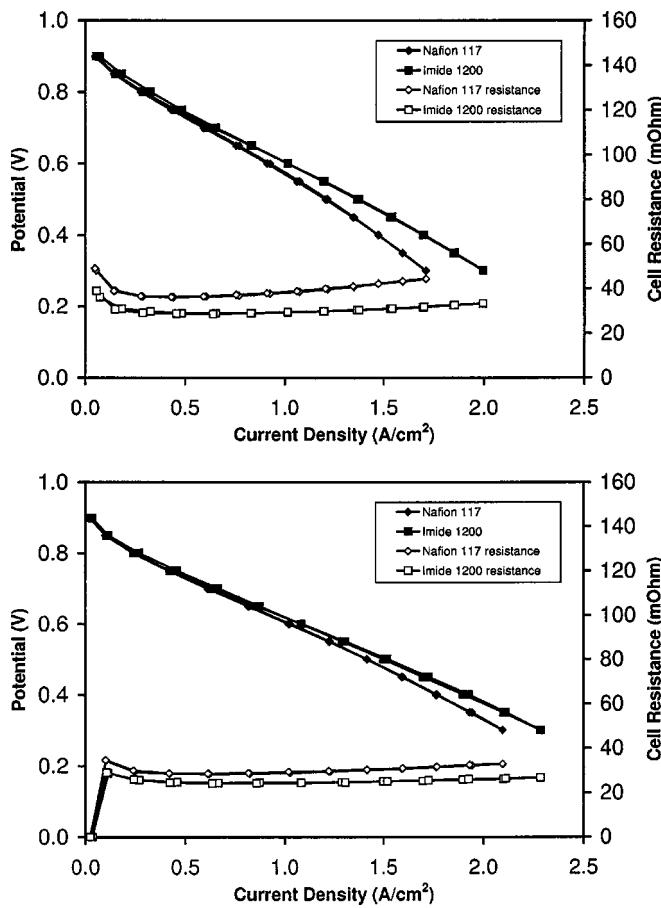


Figure 3. (◆, ■) H_2/O_2 polarization and (◇, □) cell resistance curves for 5 cm^2 MEAs with $0.3 \text{ mg Pt}/\text{cm}^2$ catalyst loading on anode and cathode: (top) cell operating temperature 80°C , hydrogen humidifier at 105°C , and oxygen humidifier at 90°C ; (bottom) cell operating temperature 110°C , hydrogen humidifier at 120°C , and oxygen humidifier at 115°C .

lent weight, the imide 1200 MEA was compared with the Nafion 117 MEA. Imide 1075 MEAs were compared with a Nafion 1135 MEA.

Figure 3 presents a series of polarization curves acquired for similarly prepared Nafion 117 and imide 1200 MEAs in a test fuel cell at operating temperatures of 80 and 110°C . The performance of the Nafion 117 MEA is similar to that reported by other researchers for similarly prepared MEAs.¹ In all cases the imide 1200 MEA exhibited a modest but consistent improvement in performance when compared with the Nafion 117 MEA. For example, at 0.3 V (the lowest voltage of the polarization curves), the imide 1200 cell current exceeded that of a similar Nafion 117 MEA by $200 \text{ mA}/\text{cm}^2$ at 80°C and by $250 \text{ mA}/\text{cm}^2$ at 110°C . Modest improvement was also seen in the MEA power curves presented in Fig. 4. At higher cell voltages (lower currents), the performance was similar for both MEAs. This improvement in performance is due in large part to the slightly lower resistance of the imide cell, as revealed by the resistance curves measured during acquisition of the polarization curves. In each case, the resistance of the imide 1200 MEA is lower than that of the Nafion 117 MEA. This behavior may be a consequence of the fact that the imide 1200 membrane is slightly thinner than the Nafion 117 membrane (150 vs. $175 \mu\text{m}$).

Differences in membrane resistance may be accounted for via “iR-corrected” polarization curves in which the cell voltage is adjusted to account for ohmic losses across the cell membrane. Figure 5 presents a series of iR-corrected polarization curves for the Nafion 117 and imide 1200 cells at the three cell temperatures that were tested in this work. Once differences in membrane resistance are

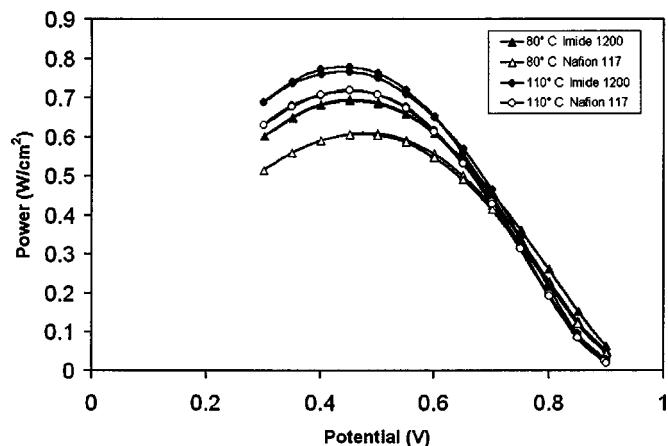


Figure 4. H_2/O_2 power curves for 5 cm^2 (Δ, \circ) Nafion 117 and (\blacktriangle, \bullet) imide 1200 fuel-cell MEAs at 80 and 110°C.

accounted for, cell performance is seen to be quite similar for both MEAs. For example, at both of the operating temperatures tested the cell performances for the Nafion and imide cells agreed to within 0.01 A/cm^2 at 0.7 V.

Figure 6 presents a series of polarization curves, and Fig. 7 presents a series of power curves acquired for similarly prepared Nafion 1135 and imide 1075 MEAs at operating temperatures of 80 and 110°C. A Nafion 1135 membrane was chosen as the benchmark in this case because it more closely matched the thickness of the two imide 1075 EW membranes. In all cases the imide 1075 MEAs outperformed the comparable Nafion 1135 MEA. The better performance of the imide 1075 MEAs is not unexpected because the imide membranes are relatively thin, which allows for better water management and maintenance of MEA hydration by water back-diffusion. While the conductivity of a imide 1075 ionomer is reported in Table I to be slightly lower than that of Nafion 1100 ionomers at 100% humidity, the imide 1075 also had significantly higher water uptake under the same conditions. Any slight deficiency in proton conductivity of imide 1075 may have been compensated by the higher water uptake, thereby yielding higher performance.

Figure 8 presents a set of iR-corrected polarization curves which to a first approximation accounts for the thickness differences between the imide 1075 and Nafion 1135 membranes (60 and 88 μm , respectively). IR-corrected performance in this case was found to be

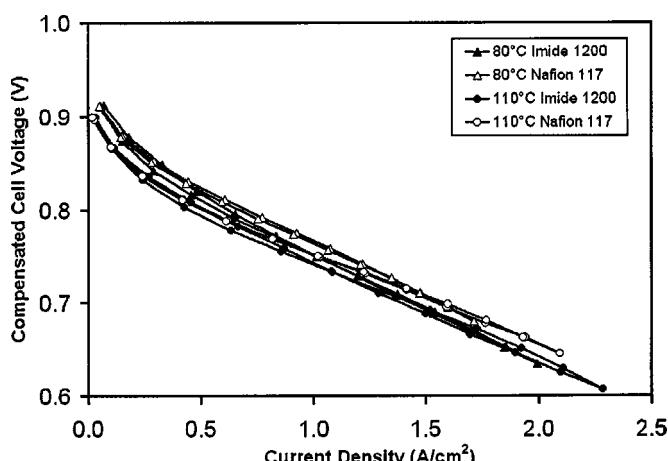


Figure 5. iR-corrected H_2/O_2 performance curves for 5 cm^2 Nafion 117 and imide 1200 fuel-cell MEAs at 80 and 110°C. Y-axis scale is restricted compared to non-iR-corrected curves to accentuate differences in performance.

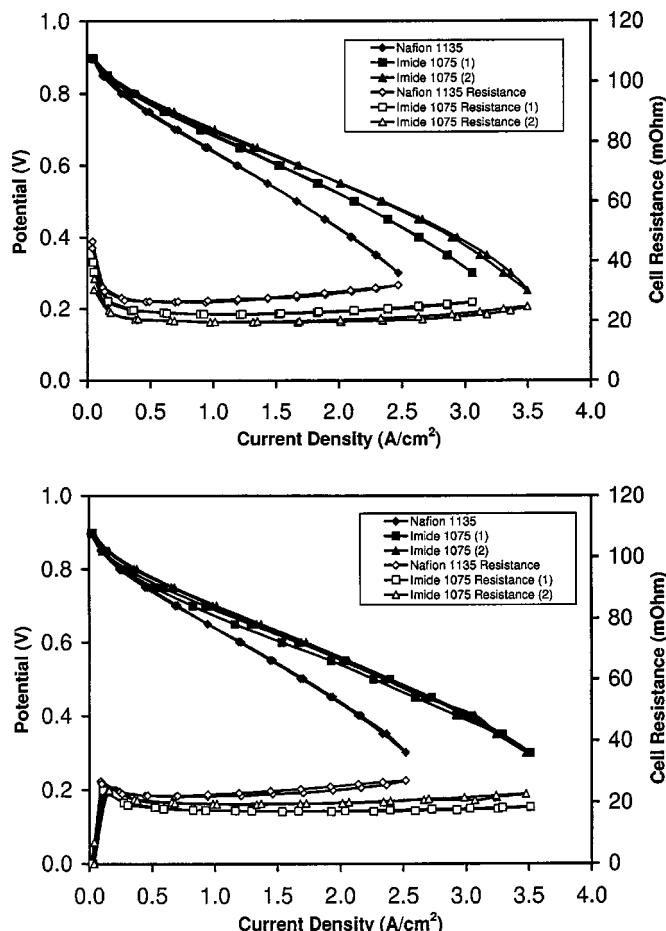


Figure 6. H_2/O_2 polarization and cell resistance curves for 5 cm^2 (\blacklozenge) Nafion 1135 and ($\blacksquare, \blacktriangle$) imide 1075 MEAs with $0.3 \text{ mg Pt}/\text{cm}^2$ catalyst loading on anode and cathode: (top) cell operating temperature 80°C, hydrogen humidifier at 105°C, and oxygen humidifier at 90°C; (bottom) cell operating temperature 110°C, hydrogen humidifier at 120°C, and oxygen humidifier at 115°C.

clustered by membrane and temperature. The Nafion 1135 MEA exhibited somewhat lower performance compared to the two imide 1075 MEAs at both temperatures. One imide 1075 MEA showed virtually temperature-independent iR-corrected performance, whereas the other imide 1075 MEA exhibited very high performance at lower temperature ($3.0 \text{ A}/\text{cm}^2$ at 0.7 V at 80°C) that diminished with an increase in temperature ($2.6 \text{ A}/\text{cm}^2$ at 0.7 V at 110°C). The good performance of the 1075 imide MEAs compared to Nafion 1135 MEAs could be a secondary effect of membrane thickness that is not fully accounted for by the iR correction, or it may reflect the favorable intrinsic properties such as high water absorption and/or better connectivity among ion-containing channels in the imide 1075 ionomers. Further work comparing MEAs that are more closely matched in thickness is required to more clearly differentiate among the possible reasons for the differing performance among these materials.

Long-term stability.—The long-term stability of the imide 1200 MEA was assessed by running the cell at 0.5 V load for nearly 600 h. Current density and cell resistance for this “life test” are shown in Fig. 9. A slow diminution in performance was observed during long-term testing of all MEAs regardless of membrane material. Workers at Los Alamos National Laboratories (LANL) reported similar losses in performance, which accounted for up to 30% of total power output within the first 500–1000 h of operation.⁸ LANL researchers attributed these losses to a slow poisoning of the plati-

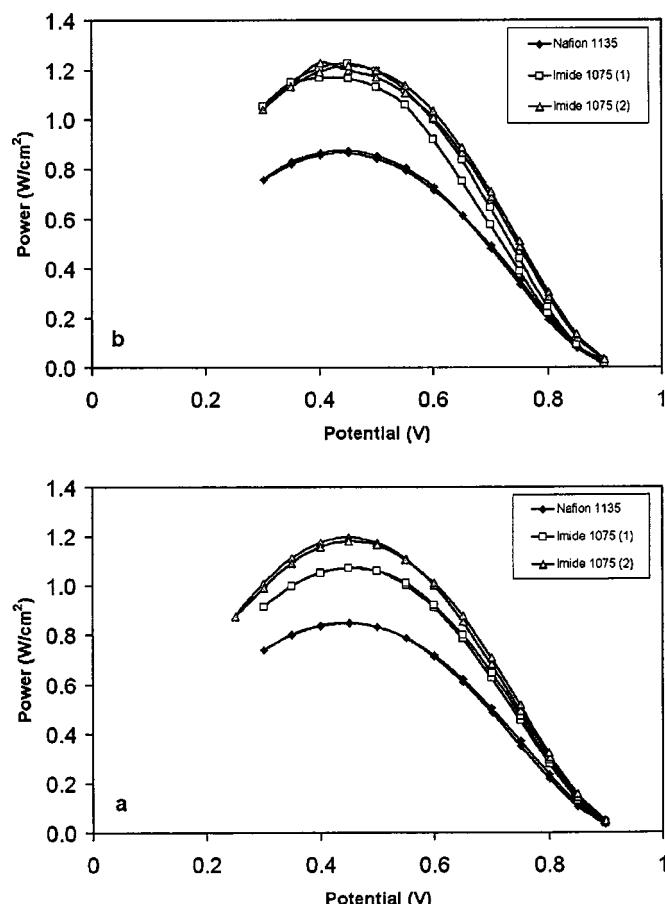


Figure 7. H_2/O_2 power curves for 5 cm^2 (◆) Nafion 1135 and (□, △) imide 1075 fuel-cell MEAs at (a) 80 and (b) 110°C.

num catalyst from low levels of CO in the hydrogen gas because injection of air into the anode reversed the effect.¹ While we cannot definitively state that the losses observed in the present work are attributable to CO poisoning, it was found that the cell resistance remained almost constant after 250 h of operation. Slow degradation of the ionomer membrane material would have likely led to a slow

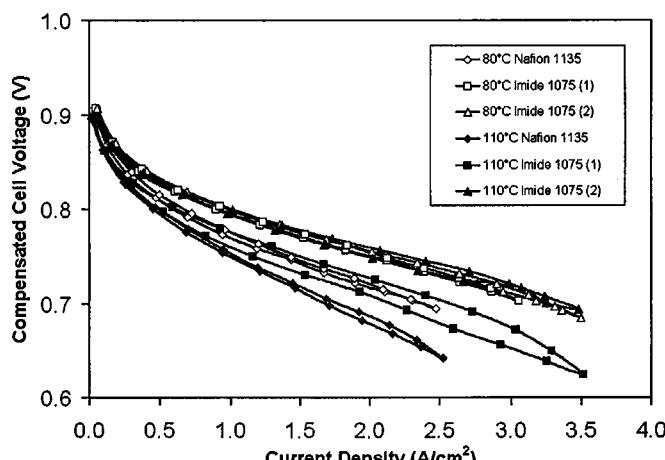


Figure 8. iR-corrected H_2/O_2 performance curves for 5 cm^2 Nafion 1135 and imide 1075 fuel-cell MEAs at 80 and 110°C. Y-axis scale is restricted to non-iR-corrected curves to accentuate differences in performance.

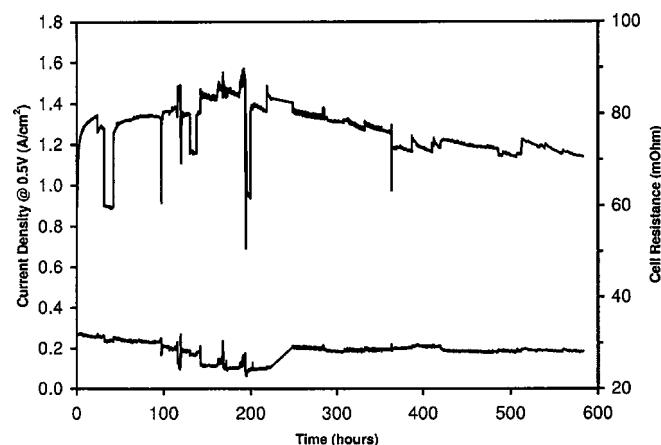


Figure 9. Long-term testing of an imide membrane fuel cell. Cell performance (top trace) and resistance (bottom trace) for 582 h of cell operation at 80°C. Hydrogen humidifier at 105°C, oxygen humidifier at 90°C. Gases supplied at four to five times stoichiometric. Both anode and cathode at 2 atm back pressure.

increase in cell resistance, which was not observed. Therefore, we believe that it is reasonable to conclude that the membrane material did not degrade significantly during the long-term testing despite the cell's slow loss of performance.

Conclusions

The performance of a test fuel cell based on a relatively thick sample of an experimental imide 1200 EW membrane is comparable to but not notably superior to that of cells based on commercially available Nafion 117 membranes. The performance of two test fuel cells fabricated using a thinner sample of an experimental imide 1075 equivalent weight (EW) membrane was notably better than that of fuel cells based on a commercially available Nafion 1135 membrane. It is considered likely that the performance of cells based on imide ionomer membranes will further improve as the techniques used to synthesize the ionomers and fabricate membranes and MEAs from them are refined. Further experiments aimed at addressing these issues are in progress.

The results presented here indicate that the effects of simply replacing the sulfonic acid functional group in Nafion ionomers with a sulfonyl imide acid group are dependent upon membrane thickness and/or EW. This is not unexpected given that conductivity and fuel-cell performance are known to be strongly dependent upon membrane thickness and ionomer EW.

The bis[(perfluoroalkyl)sulfonyl]imide motif offers opportunities for varying the ionomer primary structure and topology that are not available in sulfonic acid ionomers. Specifically, the availability of two sites on the acid group for chemical modification suggests possibilities for crosslinked ionomers and ionomers with different pendant architectures than those addressed in the present work. The present work suggests that there is a good chance that such ionomers will also exhibit good performance in PEM fuel cells, and studies aimed at synthesizing and characterizing some of these ionomers with different polymer architectures are in progress.

Finally, we note that these ionomers are still in a relatively early stage of development. No studies have yet been performed to compare the manufacturing costs of imide ionomers to other ionomers such as Nafion which are already in commercial production. At present, the imide membranes are produced through solvent casting, a technique not easily automated. However, research continues to refine both the synthesis and fabrication of these membranes, and should they continue to exhibit desirable properties, we see no fundamental reason why they should not be readily manufacturable.

Acknowledgments

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