The primary focus of this work is evaluation, modeling, and comparison of fuel cell properties for a sulfonated aromatic hydrocarbon polymer membrane under fuel cell operations against the standard commercial Nafion membrane. Membrane characterization of transport properties were reported. The fuel cell performance, current distribution mapping, and water balance measurements of both membranes were also examined at different humidity conditions. The studies show that at high RH, the hydrocarbon membrane gives better performance with less water flooding when compared with standard Nafion. However at low RH, where the membrane needs more hydration and conducting of the proton, the standard Nafion membrane shows higher performance than hydrocarbon membrane. Local distributions of current density, water content and liquid water in hydrocarbon membranes were simulated and compared with a Nafion 212 membrane. Both experimental data and model predictions show that the hydrocarbon membrane gives more uniform current distributions than that of the Nafion membrane. The computational model can predict water transport across the membrane and can be used to observe flooding in PEMFC.

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Polymer electrolyte membranes are commonly composed of a polymer backbone with negatively charged groups attached. An enhanced perfluorinated sulfonated copolymer membrane (Nafion) produced by DuPont is widely considered the standard membrane for polymer electrolyte membrane fuel cells (PEMFCs). Nafion membranes show moderate thermal and chemical stability, and have high proton conductivity under high hydration states. However, this conductivity radically decreases with temperatures higher than 90 °C because of the failure of absorbed water in the membrane. The weaknesses of this membrane to commercial use are bad conductivity at low humidity and high temperatures, chemical degradation at elevated temperatures, and the most importantly, membrane cost. The challenge presented is to produce a cheaper material for PEMFC membranes that can satisfy the thermal and chemical stability, as well as having high conductivity at low relative humidity. Currently, one of the most promising candidates for polymer backbones are sulfonated polysulfone hydrocarbon polymers. The poly(arylene ether benzonitrile) and poly(arylene ether sulfone) multiblock copolymer 6FPAEB-BPSH, which was the alternative material developed by McGrath’s research group, was used as the sulfonated polysulfone hydrocarbon in this study. Figure 1 shows the chemical structures of the 6FPAEB-BPSH multiblock copolymer (Fig. 1a) and the Nafion copolymer (Fig. 1b). Most of the hydrocarbon systems incorporate the hydrophilic “SCDPS” (sulfonated dichloro diphenyl sulfone) monomer in order to directly synthesize random copolymers or form multiblock copolymers in two steps. Research has shown the stability of these disulfonated random copolymers in accelerated studies. However, very little data has been reported on the assembly of the multiblock copolymer membranes in electrode assemblies (MEAs), and subsequent fuel cell testing for these systems. For multiblock copolymers with longer block lengths, the large amount of surface area occupied by the hydrophobic phase can lead to low connectivity at the electrode interface. Thus, the partially fluorinated hydrophobic phase in this work incorporated polar benzonitrile groups to enhance connectivity with the electrodes, while instantaneously maintaining phase separation by bringing in the fluorinated component. In fact, we previously reported good direct methanol fuel cell (DMFC) performance using a similar system. Recently, Jarrett et al. reported the investigation of multiblock disulfonated poly(arylene ether sulfone) and poly(arylene ether nitrile) copolymers. The disulfonated poly(arylene ether sulfone) and poly(arylene ether nitrile) multiblock copolymer membranes were made into membrane electrode assemblies (MEAs), and tested in different humidity environments. They concluded that the membranes vastly outperformed Nafion at high levels of relative humidity, with comparable properties to the perfluorosulfonated membrane at lower humidity.

In this work, we extend upon the work of Jarrett et al. to develop a better understanding of transport phenomena in PEMFCs. Water transport and its role in fuel cell performance is the main focus of this work. The goal is to generate fuel cell components with specific transport-dependent characteristics to enhance understanding on their impact on water management and fuel cell performance. This will be achieved by generating custom membranes (i.e., 6FPAEB-BPSH 14k-14k), characterizing them ex situ, followed by operation in a fuel cell and modeling of the results. The results are compared against a commercial membrane (i.e., Nafion 212).

**Experimental**

**Membrane characterization.** Mittelsteadt and Staser developed a dynamic approach to measuring the diffusion coefficient of water in the membranes that eliminates all inert gases (and therefore diffusion to the membrane). A detailed description of that approach is given in Ref. 18. A schematic of the apparatus is given in Figure 2. Briefly, it consists of two fast-opening pulse valves; one valve separates a dead volume from a water reservoir while the other separates the same dead volume from the chamber. This system measures dynamic water uptake in the membranes. Dynamic water absorption for both membranes were performed as follow: the valve between the membrane chamber and the dead volume was opened; the system including the chamber and the dead volume was then vacuumed and later valve opened from the vacuum pump; the valve between the chamber and the dead volume was closed to isolate these two units; a pulse valve placed between the water reservoir and the dead volume was opened allowing water vapor to enter the dead volume; finally, the
Figure 1. Structures of multiblock hexafluoropropane poly(arylene ether benzonitrile) disulfonated poly(arylene ether sulfone) copolymers and Nafion membranes. (a) Hydrocarbon and (b) Nafion.

Figure 2. Schematic of apparatus for dynamic water technique to measuring diffusion coefficient of water in the membranes.18

valve between the chamber and the dead volume was opened again allowing the membrane to absorb water vapor present in the system. The kinetics of water absorption can be monitored by the decrease in chamber pressure after the addition of water vapor, allowing the calculation of permeability. Knowing the amount of water added to the chamber also allows the calculation of water uptake, or water concentration. Calculating diffusivity from permeability and diffusivity is then trivial. In their work the authors showed that by removing gas/gas diffusion that interfacial effects were negligible and that diffusivity during adsorption or desorption, or in the steady state were identical.

To conduct the electroosmotic drag coefficient (EODC) measurement, hydrogen was fed into the anode flow field at constant pressure and excess flow through a saturator at a fixed dew point. Subatmospheric pressures were obtained by equilibrating the cell with a vacuum pump and a needle valve on the anode outlet. Vaisala humidity probes monitor the RH on both sides of the cell, and the cathode was maintained at the same humidity as the anode using a vacuum pump and a feedback controlled proportional valve. This control minimized any through plane diffusive effects. Once the cell equilibrated, a power supply applied a constant current to the cell, and the anode outlet of the cell was shut off. Constant pressure was maintained on the anode throughout the run, and the gas was humidified to a known dew point using a saturator, so the feed ratio of water molecules per proton (in the form of hydrogen gas) entering the cell could be determined from the total gas pressure. If the feed ratio was less than EODC, water molecules would be transported across the membrane more quickly than they were being fed to the anode, so the cell would dry out during the run as the water molecules on the anode that accompanied the protons across the membrane were not replaced. Cell voltage would increase along with membrane resistance until the experiment was stopped when the voltage rose above a set limit (generally 0.5 V). Likewise, if the feed ratio was greater than the EODC, water would accumulate on the surface of the anode and flood the electrode at which point the voltage would rise rapidly until it surpassed the limit.
and the experiment was stopped. Only if the feed ratio was exactly equal to the EODC would the cell operate in the metastable steady state. Runs were conducted over a range of pressures until an optimally stable configuration was found and the EODC at the given temperature and pressure was determined.

**Performance measurement.**—In this work, the catalyst used for both the anode and cathode was Tanaka Pt/C (Pt: 46.7 wt%). The Tanaka Pt/C (0.5 g) was mixed with 3.4 g of a 5 wt% commercial Nafion dispersion and 2 g of propanol. The ink mixture was subjected to ultrasonic treatment for 1 hr, and then it was sprayed onto both sides of the membranes. The note that the hydrocarbon membrane was ∼30 microns in thickness. Both the anode and cathode had a platinum loading of 0.3 mg/cm² with 25 wt% Nafion and 75 wt% Pt/C. The MEA was assembled into a single 50-cm² cell from Fuel Cell Technology. The cell hardware consisted of triple serpentine flow-field on the anode side and quadruple serpentine flow-field on the cathode side under co-current flow direction. Table I summarizes the geometry details of the hardware. In the experiments, the AvCarb series EP40T gas diffusion layer (GDL) was used for both anode and cathode. Silicone rubber-coated fiberglass fabric (SCF1007) with a thickness of 178 μm (∼7 mils) from Saint Gobain was used as a gasket for assembling the cell. All of the bolts were evenly tightened at 5.65 N-m (50 in-lb) to provide suitable compression force and prevent leaks. The experiments were performed using a Scribner 850e fuel cell test station. High purity hydrogen (99.997%) and compressed air were supplied to the anode and cathode, respectively. The fuel cell performance measurement.—The fuel cell operation by making a water balance with the inlet humidity data, the water collection data, and the fuel cell performance data. Different types of membranes differently control water transport across between anode and cathode. Figure 2 shows the experimental setup of the water balance measurement. During the performance measurement, the water in the exhausted gases was first condensed by an ice bath before entering the humidity chamber. The humidity chamber shown in Figure 3 consists of a humidity sensor and probe (Vaisala model HMT 300) with an insulated drum. Note that in some testing conditions, the outlet gas could be oversaturated and this can damage the humidity probe and sensor in the humidity chamber. Therefore, it is necessary to condense the water from the outlet gas to ensure that the gas is below 100% RH before entering the humidity chamber. The condensed water was collected and weighed every five to ten minutes over the eight to ten hours of the experiment at each cell temperature. Meanwhile, the unsaturated gas coming out from the scale (see Fig. 3) entered the humidity chamber to measure the relative humidity. This value was used to determine the amount of water in the unsaturated gas and this value was combined with the amount of condensed water from the ice bath to calculate the total water coming out of the fuel cell.

**Model Development**

A finite control volume technique based on a commercial flow solver, STAR-CD 4.18, was used to solve the coupled governing equations. This software has an add-on module called expert system of proton exchange membrane fuel cell (ES-PEMFC) version 2.50 that incorporates multi-physics of PEMFCs. These require the source terms for species transport, multiphase flow, and heat generation equations. This model has been validated against experimental data and the results were agreed for both polarization data and water balance data.

Furthermore, the predictions from numerical simulation of PEMFC were compared with segmented data taken from CDB. The model geometry used in the simulation was the same as the one used in the experiment. The geometry details for this simulation are also given in Table I. Again, the operating conditions input into the model were consistent with experimental conditions given in Table II. The

**Table I. Geometry details.**

| Description           | Value                |
|-----------------------|----------------------|
| Active area           | 50 cm²               |
| Channel width         | 0.8 mm               |
| Channel height        | 0.5 mm               |
| Rib-spacing width     | 0.8 mm               |
| GDL thickness         | 200 μm               |
| Membrane              | 6FPAEB-BPSH 14k-14k vs. Nafion 212 |

**Figure 3.** Water balance setup.
Table II. Experimental condition.

| T_{cell} (°C) | Anode RH (%) | Cathode RH (%) | Anode RH (%) | Cathode RH (%) | Back pressure (PSIG) |
|--------------|--------------|----------------|--------------|----------------|---------------------|
| 80           | 50           | 50             | 1.5          | 2.0            | 0                   |
| 80           | 95           | 95             | 1.5          | 2.0            | 0                   |
| 80           | 100          | 50             | 1.5          | 2.0            | 0                   |

Figure 4. Membrane characterization as function of relative humidity. (a) Membrane water content and (b) Proton conductivity (S/m) of membrane.

Figure 5. Membrane characterization as function of membrane water content. (a) Water diffusivity and (b) Electro-osmotic drag coefficient.

Results and Discussion

Figures 4 and 5 present the data from membrane characterization measurements described in the Experimental section. Figure 4a shows membrane water content as a function of relative humidity. The Nafton 212 membrane has higher membrane water content than the hydrocarbon membrane when relative humidity is below 65%. For RH above 65%, the hydrocarbon membrane has higher membrane water content than Nafton. The diffusion coefficients for membranes were measured over a range of membrane water contents by absorption and desorption experiments. These dynamically measured diffusion coefficients are plotted as shown in Figure 5a. It clearly presents that water diffusivity inside the membrane of Nafton is higher than hydrocarbon for the entire range of membrane water content. However, the difference between these membranes is small when the membrane water content is higher.

Figure 4b shows the proton conductivity of both the Nafton and the sulfonated polysulfone hydrocarbon membranes. Similar to the profiles of membrane water content in Figure 4a, the proton conductivity of Nafton membrane is higher than the hydrocarbon membrane with increasing relative humidity above 80 percent. Then the proton conductivity of hydrocarbon membrane becomes slightly higher than Nafton membrane. From the data of membrane water content against relative humidity shown in Figure 4a, the EODC can be plotted as a function of membrane water content as presented in Figure 5b. It shows that the Nafton membrane has higher EODC than hydrocarbon membrane and be significantly higher when the membrane water content greater.
The overall performance of PEMFCs using both Nafion and hydrocarbon membranes under three different operating conditions are shown in Figure 6. In this figure, the HFR data at three operating conditions are also reported and they all have similar profile between both of membranes. Under drier humidity condition of both anode and cathode (50/50% RH), the performance of PEMFC using the Nafion membrane is higher than the one using the hydrocarbon membrane. For this particular condition when the membrane is relatively dry, the PEMFC with the hydrocarbon membrane gives higher membrane resistance loss than PEMFC with the Nafion membrane. When the humidity condition shifts to well humidified operation (e.g., 95/95% RH), the performance of PEMFC using these two types of the membrane is opposite from what they was observed under drier conditions. The performance curve of the fuel cell using the hydrocarbon membrane increases significantly while the polarization of the cell using Nafion drops dramatically. When inlet humidity conditions change to partially dry at the cathode and the anode humidity maintains high hydration, the fuel cell performance of the membranes are similar from zero up to a current density of 1 A/cm². Higher than 1 A/cm² current density the performance of the PEMFC with Nafion drops and is significantly lower than hydrocarbon membrane. These experimental results indicate the capability of these two membranes under different humidity conditions and they both give dissimilar response according to their properties mentioned in Figs. 4 and 5.

Figure 7 presents the effect of anode and cathode relative humidity on current density at different cell potentials for both hydrocarbon (Figure 7a) and Nafion (Figures 7b) membranes. These figures intend to show that under different membrane characteristics and properties, the performance response to relative humidity is different. For PEMFC with the hydrocarbon membrane shown in Figure 7a, the current densities of three potentials (0.7V, 0.6V, and 0.5V) increase as the anode/cathode RH is increased and they are stable after the RH reaches 85%. Meanwhile, PEMFC with Nafion membrane, the current densities also rise as the anode/cathode RH is increased until the RH reaches around 75%. Then, all current densities of three set potentials drop significantly. From the experimental results given in Figures 6 and 7, it clearly confirms that Nafion membrane does not have a strong favor to be used in PEMFC under wet or high humidity operating conditions compared to the sulfonated polysulfone hydrocarbon membrane. This is because the transport properties of water for these two membranes are different as shown in Figures 4 and 5.
This figure confirms that the water flux across the membrane from the anode to the cathode inside PEMFC using Nafion membrane is much higher than hydrocarbon membrane. This data is consistent with the data of water diffusivity inside the membrane and electro-osmotic drag coefficient given in Figure 4. At the higher water flux across from anode to cathode along with water production from the electrochemical reaction, the chance of water flooding at the cathode of PEMFC using the Nafion membrane is much higher than that using the hydrocarbon membrane. However, for the case of dryer humidity condition that the water flooding is minimal, the performance of PEMFC using the Nafion membrane has much lower potential than the cell using the hydrocarbon membrane, particularly in this operating condition. This is because at higher humidity, the water transport properties of the Nafion membrane such as water diffusivity and the electro-osmotic drag coefficient are higher than the hydrocarbon membrane. This allows a higher rate of water transport to the cathode, thus high flooding occurs toward the exit of the cell. The flooding in the cell can cause the higher non-uniformity in the PEMFC current distribution with the Nafion membrane when compared to that of the PEMFC with the hydrocarbon membrane as shown in Figure 9b. This figure also shows the excellent agreement between experimental data and model predictions.

Figure 10 shows the numerical prediction of water distribution at high humidity conditions of both Nafion and hydrocarbon membranes (Figure 9a). The local current densities at ten segments using CDB measurement are also presented and compared with the numerical predictions shown in Figure 9b. Note that the flow direction and the flow-field path corresponding to each segment on the CDB can be found in Figure 8a. This figure shows overall local distribution of current density between these two membranes. Both of them have a similar distribution, in that the high current density appears at the inlet areas as shown in locations 1, 2, 3 and 4, then the current density decreases along the way toward the outlet area as presented in locations 7, 8, 9, and 10. For the PEMFC with the Nafion membrane, there is an apparent increase in experimental current density at the channel entrance, but this is attributed to the high experimental error in sections Introduction, Experimental, and Model development. The PEMFC using the Nafion membrane has much lower potential than the cell using the hydrocarbon membrane, particularly in this operating condition. This is because at higher humidity, the water transport properties of the Nafion membrane such as water diffusivity and the electro-osmotic drag coefficient are higher than the hydrocarbon membrane. This allows a higher rate of water transport to the cathode, thus high flooding occurs toward the exit of the cell. The flooding in the cell can cause the higher non-uniformity in the PEMFC current distribution with the Nafion membrane when compared to that of the PEMFC with the hydrocarbon membrane as shown in Figure 9b. This figure also shows the excellent agreement between experimental data and model predictions.

This table summarizes the water balance between the Nafion and hydrocarbon membranes at different current densities and humidity conditions.

### Table III. Water balance between Nafion and Hydrocarbon membranes.

| i A/cm² | %RH | Water in (mg/sec) | Water out (mg/sec) | Cross to Cathode (mg/sec) | Water in (mg/sec) | Gen. (mg/sec) | Water out (mg/sec) | Cross from Anode (mg/sec) |
|---------|-----|------------------|--------------------|--------------------------|------------------|--------------|-------------------|--------------------------|
| NRE     | 0.4 | 95               | 1.10               | 0.65                     | 0.46             | 3.51         | 0.93              | 4.91                     | 0.47                     |
| HC      | 0.4 | 95               | 1.10               | 0.86                     | 0.25             | 3.51         | 0.93              | 4.66                     | 0.22                     |
| NRE     | 0.6 | 95               | 1.66               | 0.96                     | 0.69             | 5.26         | 1.40              | 7.36                     | 0.70                     |
| HC      | 0.6 | 95               | 1.66               | 1.29                     | 0.37             | 5.26         | 1.40              | 7.04                     | 0.38                     |
| NRE     | 0.8 | 95               | 2.21               | 1.35                     | 0.86             | 7.03         | 1.87              | 9.80                     | 0.86                     |
| HC      | 0.8 | 95               | 2.21               | 1.68                     | 0.53             | 7.03         | 1.87              | 9.44                     | 0.54                     |
Figure 9. Numerical prediction and experimental result of current density distributions between Nafion vs. Hydrocarbon membranes. (Operating condition: co-current flow, 80°C, 1.5/2.0 stoich, H2/Air, 95/95% RH, Iavg = 1000 A/cm²). (a) Current density distribution and (b) Comparison of current density distribution between Experimental data and model prediction.

Figure 10. Numerical prediction of local distributions of water in membranes between Nafion vs. Hydrocarbon membranes. (Operating condition: co-current flow, 80°C, 1.5/2.0 stoich, H2/Air, 95/95% RH, Iavg = 1000 mA/cm²). (a) Liquid water film thickness (mm) on cathode MEA surface and (b) Membrane water content distribution.
Figures 10b presents the distributions of membrane water content for both membranes. Though water contents for both membranes are similar, the higher EODC of Nafion leads to greater flooding at the cathode. The predictions calculated by the CFD model can be seen in polarization curves given in Figure 6b and the cause of performance difference is supported by Figures 9 and 10.

Conclusions

The water content, proton conductivity, water diffusivity, and electroosmotic drag coefficient of a Nafion and hydrocarbon membranes were successfully measured and the data were included as data for model parameters. Moreover, the CDB was placed inside the fuel cell with minimal obstruction or resistance during operations for current distribution mapping. The CFD performance was verified using resistance collection. This technique was consistent with the modeled prediction.

Evaluation of fuel cell performance showed that at high RH, the hydrocarbon membrane has better performance than standard Nafion. This is because the standard Nafion membrane has higher water diffusivity and EODC than the hydrocarbon membrane, resulting in higher water flux across the membrane from anode to cathode and causing more cathode flooding in the cell. At the low RH condition where the membrane needs more hydration for better conducting of the proton, the standard Nafion membrane shows higher performance than hydrocarbon at low inlet humidity conditions. Local distributions of current density, water content and liquid water in hydrocarbon membranes were simulated and compared with Nafion membrane. The local distributions of hydrocarbon membrane show more uniformity than that of Nafion membrane. The CFD model can be used to predict water transport across the membrane and can be used to observe flooding in PEMFC.

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