Proton Conductivity of Nafion 117 as Measured by a Four-Electrode AC Impedance Method

Yoshitsugu Sone,[a] Per Ekdunge, and Daniel Simonsson*

Department of Chemical Engineering and Technology, Applied Electrochemistry, Royal Institute of Technology, S-100 44 Stockholm, Sweden

ABSTRACT

The proton conductivity of Nafion 117 was measured under various conditions of humidity and temperature using a four-electrode ac impedance method. The conductivity of this membrane without heat-treatment was *ca.* 7.8×10^{-2} S cm^{-1} at ambient temperature and 100% relative humidity; it varied strongly with the humidity and heat-treatment of the membrane. After heat-treatment, the membrane showed a slight dependence of conductivity on temperature. From 21 to 45°C, its conductivity at a given relative humidity decreased with increasing temperature, while from 45 to 80°C it increased with temperature.

Nafion 117® is a proton conducting ion-exchange membrane which is now receiving much attention due to its use as an electrolyte in the polymer-electrolyte-membrane fuel cell (PEMFC), which is of great interest for electric vehicle propulsion.[1-3] The performance of the PEMFC is strongly dependent on the humidification of the membrane, since membrane resistance changes drastically with water content. The water balance of the fuel cell is also extremely complicated. During the cell reaction in the PEMFC, the anode side (fuel side) of the membrane is readily dried up by the electro-osmotic drag of the proton, while the water produced in the cathode reaction diffuses back to the anode side. Analysis of the performance of a PEMFC requires reliable conductivity data for the membrane electrolyte at different humidities in combination with water uptake,[4-7] water diffusion,[8] gas diffusion properties,[9-14] etc.

The ionic conductivity of membranes has been measured conventionally using an ac impedance method with two electrodes.[6-8,15,16] Using this two-electrode method for a material with a low resistance, a high frequency (from 1 to 6 kHz) is needed to separate membrane resistance from interfacial capacitance, while the measurements can be practically affected by the electric fields produced by other instruments, especially in the high-frequency range. Cahan and Wainright,[17] however, reported that they used a four-electrode system for impedance measurements and successfully measured membrane impedance, which is separated from interfacial capacitance over a wide range of frequency from dc to 10^5 Hz. Deslouis and others used a four-electrode impedance method to study ionic conductivity of electronically conductive polymers,[18,19] and also mentioned the necessity of using a four-electrode arrangement to measure the very low impedance of membranes.

In this study, using this four-electrode ac impedance method, the dependence of conductivity on both relative humidity and temperature is reported. The results are discussed in terms of the extent of water uptake from the vapor phase.[4-7]

Experimental

Pretreatment of Nafion 117.—A commercial Nafion 117 membrane from Du Pont de Nemours and Company was pretreated in the conventional way described in the literature.[20-22] First, it was boiled in a 3% H_2O_2 aqueous solution for 1 h, rinsed in boiling deionized water repeatedly, boiled in 0.5 *M* H_2SO_4 for 1 h, and finally rinsed again in deionized water. After this treatment, the membrane was stored in deionized water.

Conductivity measurements.—The pretreated membrane was fixed in a cell (Fig. 1a), which consists of two platinum foil electrodes 3 cm apart to feed current to the sample (3×1 cm^2), two platinum needles 1 cm apart to measure the potential drop along the film near the center, a Plexiglas framework, and copper screws to fix these electrodes. The cell was placed in a double-mantled vessel with leads as shown in Fig. 1b, in which the thermostatic water was circulated in the jacket to keep the sample at the desired temperature. During measurements, the humidity was controlled, while the temperature of the vessel was fixed at 20, 30, 45, 60, 70, and 80°C. The humidity in this vessel was controlled by mixing water-saturated nitrogen gas with dry nitrogen gas. Both relative humidity (RH) and temperature inside the glass vessel were detected simultaneously using a sensor unit (Testo 601, Gmbh & Co.), which was placed close to the sample. This cell configuration allows the membrane to be exposed to the atmosphere directly and to respond sensitively to alteration in relative humidity.

Using the cell, the conductivity of the membrane was then measured in the longitudinal direction, and was calculated using the equation

$$\sigma = l/RS \qquad [1]$$

where σ, l, R, and S denote the ionic conductivity, distance between the reference electrodes, the resistance of the membrane, and the cross-sectional area of the membrane, respectively. To simplify the analysis, we assumed the thickness of the membrane to be 200 μm. The impedance measurements were carried out in the frequency region from 60 mHz to 6 MHz using a frequency-response analyzer (Solartron Model 1255 H. F. frequency analyzer) and potentiostat (Solartron Model 1286 electrochemical interface).

When the conductivity of the membrane without heat-treatment was measured, the water attached on the surface of the membrane was removed with filter paper just before it was positioned in the cell. The cell was then placed in the glass vessel, and the sample was dried using a stream of dry nitrogen gas. After the measurements under various humidification conditions and temperatures, this membrane was heat-treated at normal atmospheric pressure for 10 h. During this heat-treatment, the membrane was still tightly fixed to the cell to avoid change in the position of the electrodes.

Immediately after heat-treatment, the membrane was set in the glass vessel into which the humid nitrogen gas stream with *ca.* 80% RH was supplied. After a constant conductivity was obtained, the membrane was exposed to dry nitrogen gas, and then the conductivity measurements under various humidification conditions and temperatures were begun; relative humidity was varied in step sizes of 5 to 10%. When we increased the humidity, we observed a change in the conductivity, at a constant frequency of 100 Hz, for more than 1 h. It took about 30 min to reach a constant conductivity with increasing humidity, while it took several hours with decreasing humidity. When a constant conductivity had been obtained, it was measured in the

* Electrochemical Society Active Member.
[a] On leave from Institute of Industrial Science, University of Tokyo, 7-22-1 Roppongi, Minato-ku, Tokyo 106, Japan.

Downloaded on 2016-12-15 to IP 128.252.67.66 address. Redistribution subject to ECS terms of use (see ecsdl.org/site/terms_use) unless CC License in place (see abstract).

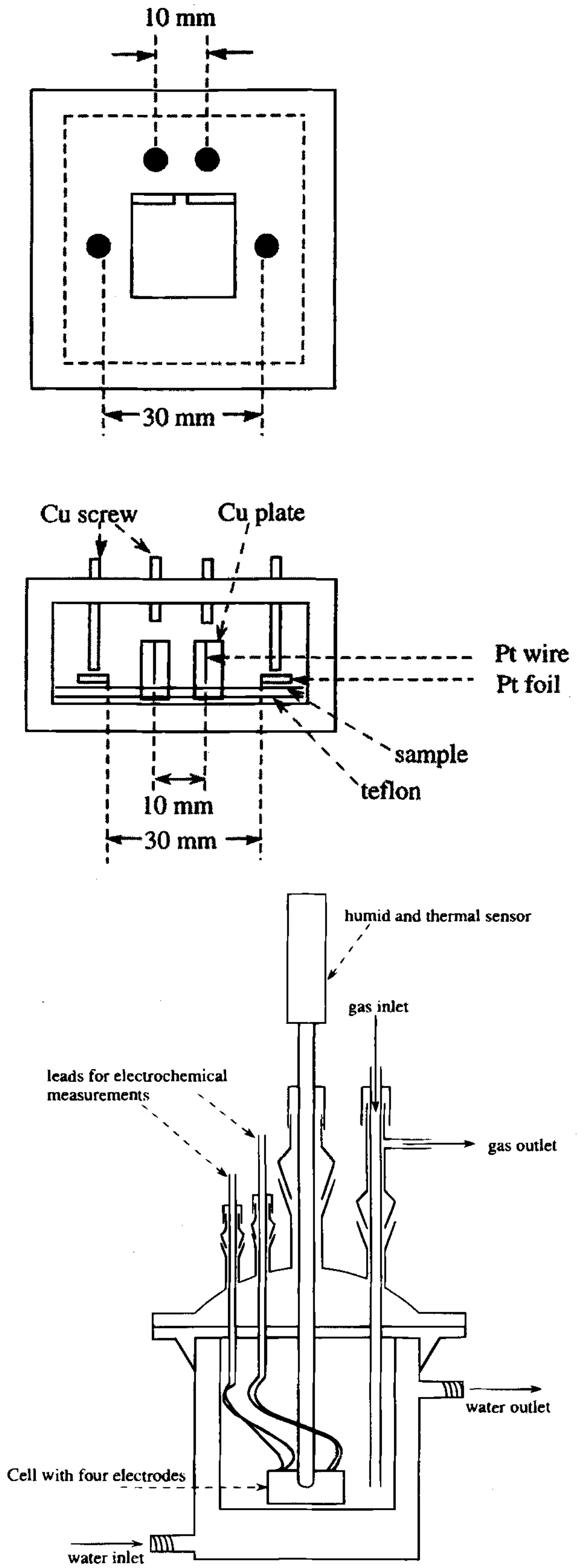

Fig. 1. Apparatus for measuring proton conductivity under exposure to a stream of humid gas. (a, top) Cell with four platinum electrodes and (b, bottom) glass vessel in which to set the cell folder.

frequency region from 60 mHz to 6 MHz. The conductivity of the membrane was thus measured after heat-treatments at 80, 105, and 120°C.

It was difficult in practice to obtain the same conductivity for different membranes. To compare the dependence of conductivity on temperature and humidity, the measurements were thus made on the same membrane. Thereafter, we checked the conductivity again using different membranes and observed the same dependence.

Results and Discussion

Impedance measurements.—Figure 2 shows a typical example obtained by the ac impedance method using four electrodes; this was obtained using the membrane without heat-treatment under 100% RH. As shown in Fig. 2, we observed an impedance independent of frequency over the range from 10^0 to 10^5 Hz. This meant we were able to separate membrane resistance from interfacial resistance in this frequency region. Using a Bode plot, we first checked the frequency region over which the impedance had a constant value. The resistance of the membrane was then obtained from a Cole-Cole plot. For instance, the conductivity calculated in Fig. 2 is 7.8×10^{-2} S cm^{-1}, almost identical with that calculated from two-electrode impedance measurement using a swollen membrane.[6,7,15] When the two-electrode system was used, the resistance was usually measured in the tranverse direction (*i.e.*, across the membrane as in a PEMFC). In Nafion, protons migrate via water molecules absorbed in the micropores. Since Nafion has an amorphous structure with micropores, it is reasonable to expect an isotropic proton movement which is independent of the direction of the measurement.

Basic proton conductivity of Nafion 117.—Conductivity measurements were carried out using a membrane without heat-treatment and with heat-treatment at 80, 105, and 120°C. Just after the heat-treatment, the membrane performed like an insulator. Figure 3 shows the change in

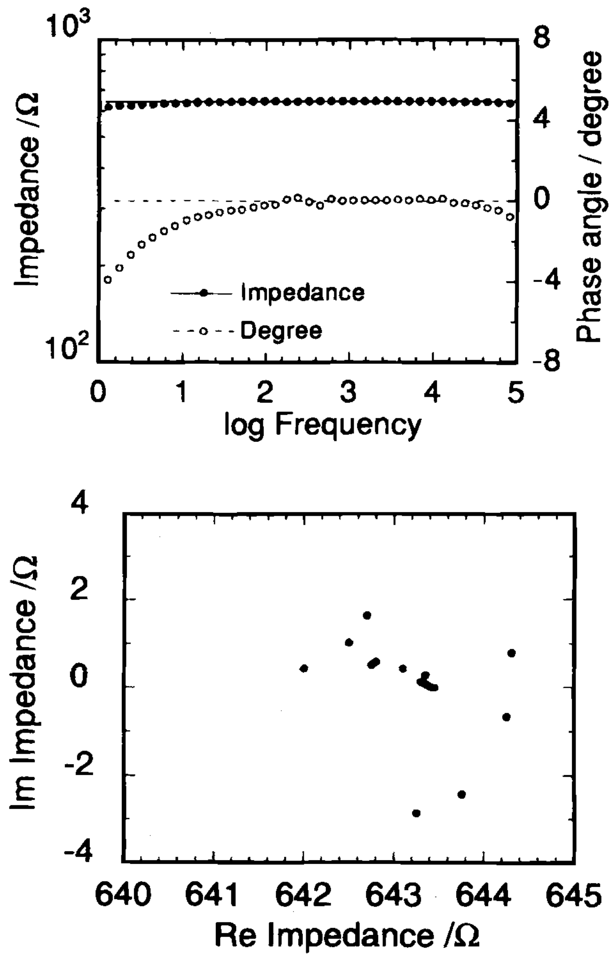

Fig. 2. Typical expression of impedance analysis using four-electrode impedance method, using (a, top) Bode type plot and (b, bottom) Cole-Cole type plot.

Downloaded on 2016-12-15 to IP 128.252.67.66 address. Redistribution subject to ECS terms of use (see ecsdl.org/site/terms_use) unless CC License in place (see abstract).

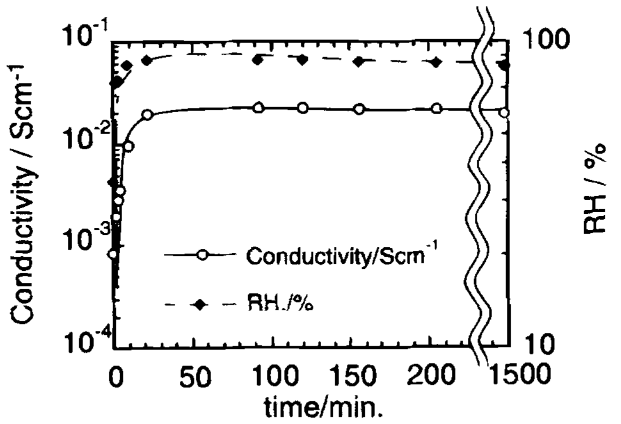

Fig. 3. Change in conductivity after the heat-treatment at 80°C under an RH of 80%.

conductivity of the membrane after heat-treatment at 80°C. When this membrane was first placed in the gas stream and the relative humidity varied up to *ca.* 80%, the conductivity increased rapidly with relative humidity and reached a constant value after 30 min. We maintained this membrane under the same conditions for 24 h and obtained almost the same conductivity. After heat-treatment at 105 and 120°C, we observed the same type of change in conductivity when the membrane was first exposed to a humid condition.

Figure 4 shows the change in membrane conductivity with increasing heat-treatment temperature. The change in membrane structure and water uptake performance after heat-treatment was reported by Yeo and Yeager,[23] who described three different forms of the membrane. The membrane without heat-treatments was called E-form (expanded form), and the membranes which were heat-treated at 80 and 105°C were N-form (normal form) and S-form (shrunken form), respectively. According to a previous study, the membrane is supposed to have a porous structure.[24] When the membrane is heat-treated at 80°C, some of the micropores are attached to each other, and some are closed by the thermal treatment. When the membrane is heat-treated at 105°C, the shrinkage of the membrane further increased. This is because the Nafion-type of membrane has a glass-transition temperature of about 110°C, at which value molecular motions take place more easily.[25] This molecular motion changes the structure of the membrane and causes a lower water content of the S-form than of E- and N-forms. According to Ref. 4, the amount of water uptake from liquid water decreases from

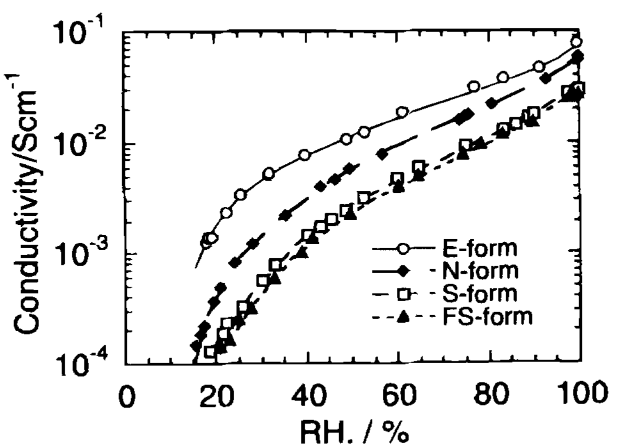

Fig. 4. Conductivity of Nafion 117 using the membrane (O) without heat-treatment (E-form), with heat-treatment at (◆) 80°C (N-form), (□) 105°C (S-form), and (▲) 120°C (FS-form).

0.37 (E-form) to 0.22 (N-form) and to 0.18 g H_2O/g dry membrane (S-form). The amount of water absorbed from the vapor phase is even less than that from the liquid phase, and also decreases with increasing heat-treatment temperature.[4] Furthermore when the proton conductivity is measured, protons located near the sulfonic groups migrate via water molecules. Thus a lower water content of the membranes results in a decrease of proton carriers, and hence a lower proton conductivity. The membrane heat-treated at 120°C shows a lower conductivity than that heat-treated at other temperatures (E-form, N-form, and S-form). As stated above, Nafion has a glass-transition temperature of 110°C. The fuel-cell electrodes are normally hot-pressed to the membrane at still higher temperatures. The difference in the conductivity at 105 and 120°C indicates that the membrane shrinks further (FS-form) at the higher temperature; the difference in the conductivity is, however, very small. For a further analysis, the extent of water uptake from vapor phase is needed.

Dependence of conductivity on relative humidity and temperature.—Figure 5 shows the dependence of the conductivity of Nafion 117 on humidity and temperature. The conductivity of this membrane depends strongly on humidity and slightly on temperature. The measurement at 80°C using the E-form of the membrane was not possible since, at this temperature, the conductivity of the E-form decreased rapidly to that of the N-form.

The temperature dependence of conductivity can be described as follows. When the conductivity was measured at 20, 30, and 45°C at the same value of RH, it decreased with increase in temperature as shown in Fig. 5a, c, e, and g. It then increased when the temperature was increased from 45 to 80°C as shown in Fig. 5b, d, f, and h. The relationship between ionic conductivity and temperature can be expressed by the Arrhenius relation

$$\sigma = A \exp(-E_a/RT) \qquad [2]$$

where σ, A, E_a, R, and T denote the ionic conductivity, frequency factor, activation energy for ionic conduction, gas constant, and temperature, respectively. This equation indicates that the conductivity of the membrane should increase when the temperature of the sample increases. In contrast, the series of results obtained here at 20, 30, and 45°C could not thus be explained using the above equation.

The water uptake of the membrane from the vapor phase at different temperatures has been measured in our laboratory with RH fixed at 100%. The water uptake decreased drastically between 20 and 45°C, while above 45°C it was rather constant. Comparing these results to those obtained in the conductivity measurement, we can say that the decrease of conductivity between 20 and 45°C was due to the decrease of water content in the membrane. Above 45°C the membrane had a fairly constant amount of water, and the increase in conductivity with temperature from 45 to 80°C can be explained on the basis of the Arrhenius equation. Analysis of the water uptake measurements will be made elsewhere.

References 4 and 5 reported water uptake of membranes from the atmosphere. In these reports, polynomial equations of order three were fit to the relationship between relative humidity and membrane water content. To our knowledge, no scientific reason has been shown for the order of this polynomial equation. The curves in Fig. 5 could also be well fit using the following equation, especially under high-humidity conditions (RH = 40 to 100%), though we have no scientific reason yet for this order

$$\sigma = a + bx + cx^2 + dx^3 \qquad [3]$$

where σ and x denote ionic conductivity and relative humidity, respectively, and the coefficients of a, b, c, and d are listed in Table I.

According to Eq. 3 and Table I, the conductivity at 100% RH was calculated at temperatures of 45, 60, 70, and 80°C. Using these results, the activation energies for proton conduction, according to Eq. 2, were calculated and are shown

Downloaded on 2016-12-15 to IP 128.252.67.66 address. Redistribution subject to ECS terms of use (see ecsdl.org/site/terms_use) unless CC License in place (see abstract).

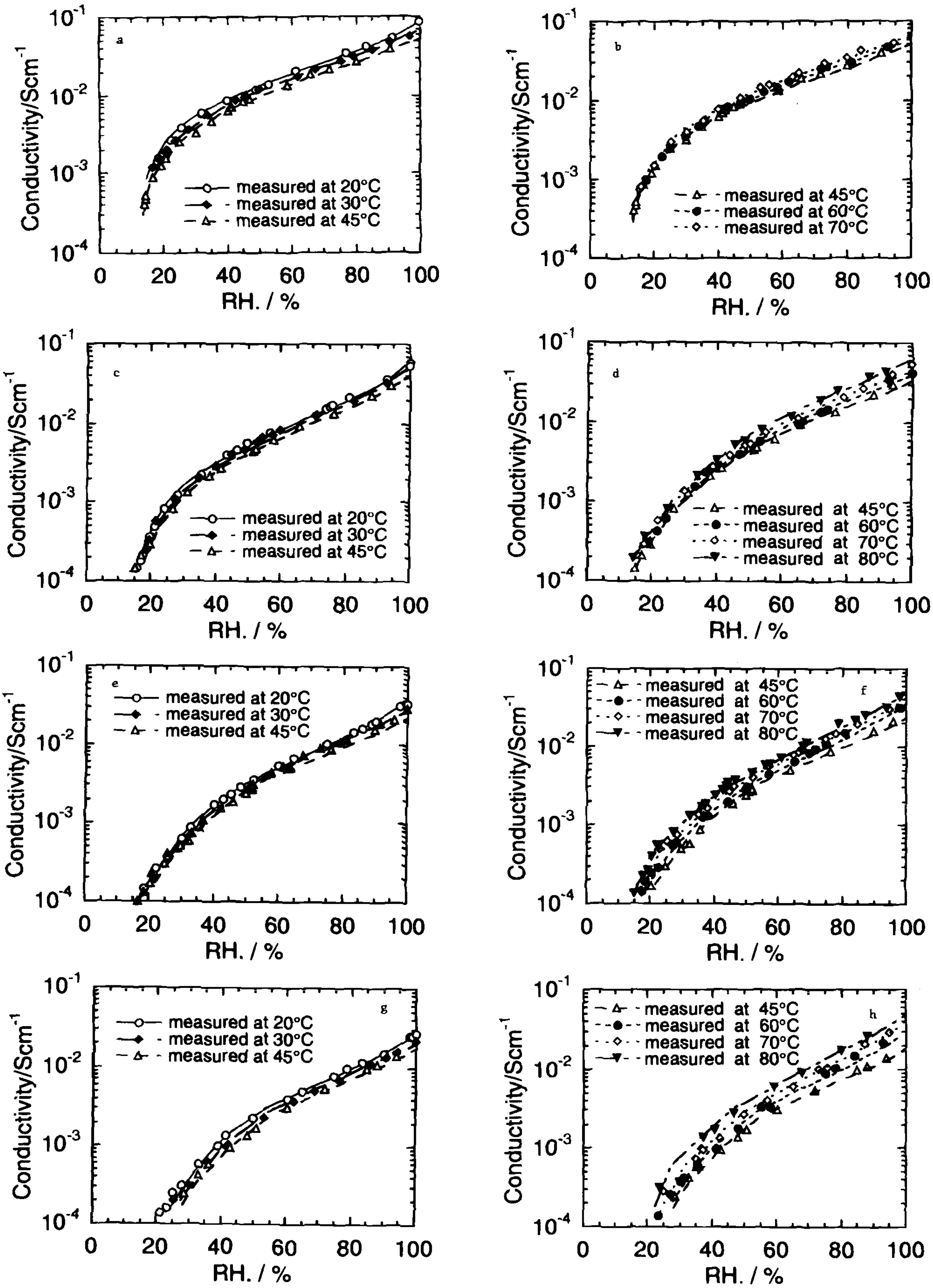

Fig. 5. Conductivity dependence on temperature and relative humidity. (a) Conductivity measured at 20, 30, and 45°C using the membrane of the E-form. (b) Conductivity measured at 45, 60, and 70°C using the membrane of the E-form. (c) Conductivity measured at 20, 30, and 45°C using the membrane of the N-form. (d) Conductivity measured at 45, 60, 70, and 80°C using the membrane of the N-form. (e) Conductivity measured at 20, 30, and 45°C using the membrane of the S-form. (f) Conductivity measured at 45, 60, 70, and 80°C using the membrane of the S-form. (g) Conductivity measured at 20, 30, and 45°C using the membrane of the FS-form. (h) Conductivity measured at 45, 60, 70, and 80°C using the membrane of the FS-form.

Downloaded on 2016-12-15 to IP 128.252.67.66 address. Redistribution subject to ECS terms of use (see ecsdl.org/site/terms_use) unless CC License in place (see abstract).

Table I. Coefficients for the polynomial Eq. 3.

Structure of the membrane	Temperature for measurement (°C)	a (10^{-3} S cm^{-1})	b (10^{-4} S cm^{-1})	c (10^{-6} S cm^{-1})	d (10^{-8} S cm^{-1})
E-form	20[a]	−19.8	16.6	−34.5	28.4
	30	−8.01	6.72	−11.6	11.8
	45	−1.75	1.45	0.0161	3.45
	60	−3.41	2.73	−2.67	5.72
	70	−1.56	1.21	1.01	3.95
N-form	20[a]	−11.2	9.73	−22.9	19.7
	30[a]	−5.74	4.58	−9.80	9.81
	45	−3.18	2.80	−6.55	7.63
	60	−2.84	2.51	−6.17	8.18
	70[a]	−8.36	7.35	−17.5	16.3
	80	−1.45	1.57	−4.55	8.86
S-form	20[a]	−7.52	6.23	−14.6	12.4
	30[a]	−3.78	3.13	−7.45	7.46
	45	−3.89	2.87	−6.42	6.31
	60	−3.65	3.24	−8.32	8.77
	70	−4.62	3.78	−9.02	9.34
	80[a]	−4.66	4.13	−10.3	11.0
FS-form	20[a]	−7.59	5.61	−12.4	10.2
	30[a]	−3.35	2.72	−6.53	6.20
	45	−2.58	1.74	−3.87	4.11
	60	−3.80	2.92	−7.36	7.65
	70	−7.02	5.23	−12.5	11.5
	80	−7.49	5.82	−14.1	13.4

[a] Polynomial Eq. 3 does not fit specially at RH less than 40%.

in Table II. The activation energy was less than 2 kJ mol^{-1} for all forms of membranes.

In this type of low-temperature proton conductor, proton migration is primarily by the Grothuss mechanism. In this mechanism, the proton which forms an H_3O^+ ion jumps to the neighboring lone pair of electrons of a water molecule. For such a mechanism, the activation energy for proton conduction should be about 14 to 40 kJ mol^{-1};[26] however, the values obtained in this work are much smaller than those reported in Ref. 6, 15, and 16. One explanation could be that the membrane in our measurement was fully exposed to the atmosphere. At higher temperature, the membrane keeps less water, and this decrease in water content causes a lower conductivity. On the contrary, according to Eq. 2, the conductivity increases with temperature. These two phenomena simultaneously influence the membrane conductivity and thus yield an apparent activation energy of less than 2 kJ mol^{-1} for proton conduction. The relatively small values of the activation energies suggest a Grothuss mechanism for proton conduction in Nafion 117.

As noted above, the proton conductivity depends highly on the relative humidity. For a relative humidity of less than 20%, the conductivity decreased drastically. Further, with an RH of less than 15%, Nafion 117 behaved like an insulator, indicating the absence of proton migration in Nafion without enough hydration. Using the data on water uptake from the vapor phase reported in Ref. 4-7, the proton conductivity was plotted *vs.* water content (λ = mol H_2O/mol SO_3H) as shown in Fig. 6. The value for λ was calculated for various levels of humidity conditions, using the equations shown in Ref. 4 and 5. Although the proton conductivity was supposed to be linearly proportional to the water content of the membrane,[7] two different regions are observed in Fig. 6. Between $\lambda = 2$ and 4, the proton conductivity increased exponentially; when the water content was higher than $\lambda = 5$, it increased linearly with water content. Moreover, this figure indicates that at least two water molecules per sulfonic unit are required for proton conduction.

This analysis suggests the following mechanism for water uptake. When λ is 2, the hydrated water can be divided into two groups; (*i*) water molecules are strongly localized near the sulfonic group because of strong hydrogen bonding. At the same time, these molecules do not permit other water molecules to localize near the sulfonic groups; (*ii*) water molecules, attached to the membrane via another water molecule, are held together by a weak hydrogen bond. The results obtained here indicate that this type of loosely attached water molecules is essential for proton migration. This is the reason that proton conductivity is detected only above $\lambda = 2$. With water absorption being more than $\lambda = 2$, the migration channels for protons are connected to each other, and thus the proton conduction rapidly increases with water content. This phenomenon could thus be described by a percolation mechanism.[27] Further, as previously reported, a proton drags water molecules by electro-osmotic drag during its migration;[6,7] the number dragged is 1 to 3 per proton. Including the water localized strongly on the sulfonic unit, four water molecules are needed for the best proton migration in the membrane. This number coincides with the initial value of the linear increase of proton conductivity.

Conclusion

Proton conductivity of Nafion 117 was measured before and after heat-treatment at 80, 105, and 120°C, respectively. The proton conductivity of the membrane without heat-treatment was *ca.* 0.09 S cm^{-1} at RH = 100%. Heat-treatment of membrane decreases the conductivity due to the change in its structure.

After heat-treatment, the dependence of conductivity on relative humidity was measured. This measurement revealed the strong dependence of conductivity on relative humidity. Although this dependence had been expected, such accurate measurements had not been realized previously because a four-electrode impedance measurement was not made. At least two water molecules per sulfonic unit are essential for proton conduction, and, between $\lambda = 2$ and 4, the proton conductivity increases exponentially with increase of water content. Above $\lambda = 4$, this relationship is linear.

The conductivity of the membrane decreases with increase of temperature from 20 to 45°C due to loss of water from the membrane at the higher temperature. In contrast, above 45°C the conductivity increases with temperature because the water content in the membrane remains rather constant. The activation energy obtained above 45°C was less than 2 kJ mol^{-1}.

Finally, we should mention that the proton conductivity of Nafion heat-treated at 105 and 120°C offers important parameters from a practical viewpoint. This is because, when PEMFC is fabricated, the membrane is usually hot-pressed with platinum electrodes at a temperature higher than 100°C; Nafion has a glass-transition temperature around this temperature. In addition, the

Downloaded on 2016-12-15 to IP 128.252.67.66 address. Redistribution subject to ECS terms of use (see ecsdl.org/site/terms_use) unless CC License in place (see abstract).

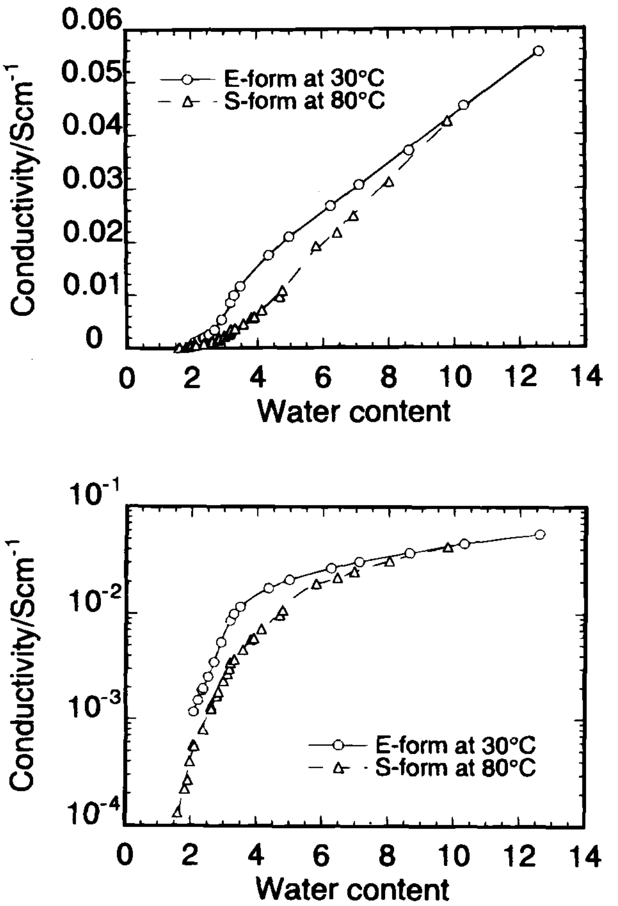

Fig. 6. Dependence of conductivity on water content, (λ = mol H_2O/mol SO_3H) of the membrane expressed using (a, top) linear scale and (b, bottom) log scale on *y*-axis.

loading temperature of PEMFC is normally 80°C. Thus, the data we obtained, *i.e.*, the dependence of conductivity on relative humidity after heat-treatment at 105 and 120°C, is important for the mathematical modeling of a PEMFC performance.

Manuscript submitted July 3, 1995; revised manuscript received Dec. 28, 1995.

Table II. Activation energy for proton migration calculated at RH = 100%.

Membrane structure	E-form	N-form	S-form	FS-form
Heat-treated temperature (°C)	—	80	105	120
Activation energy (E_a) (kJ mol^{-1})	1.1	1.3	1.1	1.8

Royal Institute of Technology assisted in meeting the publication costs of this article.

REFERENCES

1. K. B. Prater, *J. Power Sources*, **51,** 129 (1994).
2. J. P. Shoesmith, R. D. Collins, M. J. Oakley, and D. K. Stevenson, *ibid.*, **49,** 129 (1994).
3. K. Straβer, *B. Bunsenges. Phys. Chem.*, **94,** 1000 (1990).
4. J. T. Hinatsu, M. Mizuhata, and H. Takenaka, *This Journal*, **141,** 1493 (1994).
5. T. E. Springer, T. A. Zawodzinski, and S. Gottesfeld, *ibid.*, **138,** 2334 (1991).
6. T. A. Zawodzinski, Jr., C. Derouin, S. Radzinski, R. J. Sherman, V. T. Smith, T. E. Springer, and S. Gottesfeld, *ibid.*, **140,** 1041 (1993).
7. T. A. Zawodzinski, Jr., T. E. Springer, J. Davey, R. Jestel, C. Lopez, J. Valerio, and S. Gottesfeld, *ibid.*, **140,** 1981 (1993).
8. T. A. Zawodzinski, Jr., M. Neeman, L. O. Sillerud, and S. Gottesfeld, *J. Phys. Chem.*, **95,** 6040 (1991).
9. J. S. Chiou and D. R. Paul, *Ind. Eng. Chem. Res.*, **27,** 2161 (1988).
10. Z. Ogumi, Z. Takehara, and S. Yoshizawa, *This Journal*, **131,** 769 (1984).
11. Z. Ogumi, T. Kuroe, and Z. Takehara, *ibid.*, **132,** 2601 (1985).
12. T. Sakai, H. Takenaka, and E. Torikai, *ibid.*, **133,** 88 (1986).
13. M. C. Kimble, R. E. White, Y-M. Tsou, and R. N. Beaver, *ibid.*, **137,** 2510 (1990).
14. T. Sakai, H. Takenaka, N. Wakabayasi, Y. Kawami, and E. Torikai, *ibid.*, **132,** 1328 (1985).
15. J. Halim, F. N. Büchi, O. Haas, M. Stamm, and G. G. Scherer, *Electrochim. Acta*, **39,** 1303 (1994).
16. K. Uosaki, K. Okazaki, and H. Kita, *J. Electroanal. Chem.*, **287,** 163 (1990).
17. B. D. Cahan and J. S. Wainright, *This Journal*, **140,** L185 (1993).
18. C. Deslouis, M. M. Musiani, and B. Tribollet, *J. Phys. Chem.*, **98,** 2936 (1994).
19. C. Deslouis, M. M. Musiani, B. Tribollet, and M. A. Vorotyntsev, *This Journal*, **142,** 1902 (1995).
20. E. A. Ticianelli, C. R. Derouin, A. Redondo, and S. Srinivasan, *ibid.*, **135,** 2209 (1988).
21. A. Parthasarathy, S. Srinivasan, A. J. Appleby, and C. R. Martin, *J. Electroanal. Chem.*, **339,** 101 (1992).
22. S. Mukerjee and S. Srinivasan, *ibid.*, **357,** 201 (1993).
23. R. S. Yeo and H. L. Yeager, *Modern Aspects of Electrochemistry*, Vol. 16, B. E. Conway, R. E. White, and J. O'M. Bockris, Editors, p. 437, Plenum Press, New York (1985).
24. M. W. Verbrugge and R. F. Hill, *This Journal*, **137,** 3770 (1990).
25. S. C. Yeo and A. Eisenberg, *J. Appl. Polym. Sci.*, **21,** 875 (1977).
26. P. Colomban and A. Novak, *Proton Conductors*, P. Colomban, Editor, pp. 46-51, Cambridge University Press, Cambridge (1992).
27. T. D. Gierke and W. Y. Hsu, *Perfluorinated Ionomer Membranes*, A. Eisenberg and H. L. Yeager, Editors, p. 283, American Chemical Society, Washington, DC (1982).

Downloaded on 2016-12-15 to IP 128.252.67.66 address. Redistribution subject to ECS terms of use (see ecsdl.org/site/terms_use) unless CC License in place (see abstract).