FULL PAPER

Preparation procedure for the electrode slurries of polymer electrolyte fuel cells utilizing the irreversibility of ionomer adsorption onto Pt-C particles

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The effect of slurry preparation for polymer electrolyte fuel cells on the electrode performance has been investigated in this work. The amount of ionomer adsorbed onto Pt-C powder increases when water is used as a solvent; however, the size of non-adsorbed ionomer particles increases as well, resulting in a larger gas permeation resistance. Meanwhile, increasing the ethanol-to-water mass ratio in a mixed solvent decreases the ionomer size. Therefore, an improved two-step slurry preparation procedure utilizing the irreversibility of the ionomer adsorption onto Pt-C powder has been investigated. In this method, (1) Pt-C powder is mixed with ionomer in water to maximize its adsorbed fraction, after which (2) ethanol is added to the prepared slurry to minimize the size of non-adsorbed ionomer. As compared with the conventional one-step synthesis method, the developed two-step preparation procedure enhances the electrode performance without changing its composition.

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1. Introduction

Fuel cells represent a promising solution for various global environmental issues as a clean energy source. Multiple research studies have been devoted to fuel cells, and the development of fuel cell vehicles is currently ongoing. Fuel cells are classified into several types, among which polymer electrolyte fuel cells (PEFCs) are expected to become most popular in the future due to their small size, light weight, low operating temperature, and short start-up time.¹⁾ Previous works on PEFCs were mainly focused on the synthesis of catalyst powders such as Pt-BC,²⁾ Pt-Al₂O₃,³⁾ Pt nanowires,⁴⁾ and Pt-C nanoparticles.⁵⁾ On the other hand, Wang et al. investigated the effect of carbon addition to the electrode slurry on the fuel cell performance.⁶⁾ In addition, optimizations of the gas diffusion layer,⁷⁾ polyelectrolyte film,⁸⁾ ionomer type,⁹⁾ and ionomer mixing method¹⁰⁾ were conducted. However, the use of Pt catalyst most strongly enhanced the electrode performance;^{11)–13)} therefore, the optimal strategy should be aimed at reducing the amount of utilized Pt to minimize the cost of PEFCs while maintaining their superior characteristics.

For this purpose, preparation conditions for the electrode slurry during the fabrication of PEFC electrodes must be investigated. However, the number of studies on this topic is smaller than that of the reports about the synthesis of catalyst powders and PEFC raw materials. Furthermore, the majority of these studies focused on the relationship between the raw material type and fuel cell performance characteristics (such as the *I-V* curve) and not on the slurry parameters.^{14)–19)} Although the dispersion properties of catalyst powders and ionomers were investigated in some works, the relationship between the microstructure, proton conductive resistance, and oxygen transfer resistance of the electrode and the slurry properties have not been discussed in detail.^{20)–26)} Park et al. conducted the analysis of the Pt distribution and ionomer coverage on the base material powder, but they did not perform any slurry characterization.²⁷⁾ Takahashi et al. examined the relationship between the cell performance and microstructure of the freeze-dried slurry by varying the solvent type; however, the microscopic observation of the freeze-dried slurry by scanning electron microscopy (SEM) was unable to determine its macroscopic characteristics.²⁸⁾

Among these works, several studies were devoted to the effect of the slurry parameters on the cell performance. Thus, Komoda et al. examined the relationship between

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the particle dispersion in the slurry and cell properties.²⁹⁾ In their work, the slurry was characterized from the rheological viewpoint by measuring its apparent viscosity at various mixing times; however, the packing ability of the slurry was not evaluated. Nam et al. reported that the slurry formation process was more efficient than the powder mixing process in dry air; nevertheless, the optimum slurry preparation conditions were not determined.³⁰⁾ Yang et al. found that ethylene glycol was a suitable solvent for slurry preparation that significantly improved the PEFC performance, but its optimum mixing conditions were not established.³¹⁾ Similarly, Sung et al. reported that *N,N*-dimethylacetamide could be used as a solvent; however, the resulting solution was not universal and required a specific binder, and the optimal slurry preparation conditions were not determined.³²⁾ Finally, Ren et al. investigated the effect of slurry composition on the cell performance, but did not examine possible preparation methods and their conditions.³³⁾

It is well known that PEFC performance depends on the electrode microstructure. For this reason, cell performance can be controlled by varying the particle dispersion state, amount of ionomer adsorbed by the slurry powder, and slurry packing ability. In our previous work,³⁴⁾ we investigated the relationship between the cell performance and slurry characteristics including not only its rheological properties, but also the packing ability. It was found that the latter parameter as well as the amount of adsorbed ionomer and size of non-adsorbed ionomer species strongly affected the cell properties.³⁴⁾ In the same study,³⁴⁾ we suggested that the cell performance could be further improved by simultaneously increasing the amount of adsorbed ionomer and decreasing the size of non-adsorbed ionomer. It was also shown that the first parameter was relatively large when water was used as a solvent and that the ionomer size decreased with an increase in the ethanol mass ratio in a water-ethanol mixture.³⁴⁾ Therefore, the objective of this work was to examine a two-step preparation method, in which slurry was prepared in water first followed by the addition of an appropriate amount of ethanol. The characteristics of the resulting slurry were compared with those of the slurry prepared by a one-step method, and the performances of their corresponding cells were investigated. By comparing the results obtained by the one-step and two-step preparation methods, the possibility of improving the cell performance without changing the slurry composition, i.e. by increasing the amounts of added Pt (catalysts powder) and ionomer was explored.

2. Materials and methods

2.1 Materials

All materials used in this study were identical to those utilized in a previous work.³⁴⁾ Commercial Pt-C powder (TEC10V30E, 30.1 mass %-Pt, Tanaka Kikinzoku Kogyo, Japan) and Nafion (DE1021, Dupont, USA) were used as the catalyst and ionomer, respectively. Deionized water and water-ethanol mixtures with ethanol concentrations of 7.5, 15, 22.5, and 30 mass % served as the solvents.

2.2 Slurry preparation

Slurries were prepared by the one-step and two-step methods illustrated in **Fig. 1**. Their compositions are summarized in **Table 1**. In the one-step method, specified amounts of Pt-C powder, ionomer, and solvent were placed into a plastic pot and then mixed using a rotating and revolving mixer (Awatori Rentaro ARE-300, Thinky, Japan) at a rotation speed of 2000 rpm and revolution speed of 800 rpm for 15 min. In the two-step method, the aqueous slurry without ethanol with the composition listed in Table 1 was prepared first (see **Table 2**, procedure 1). Appropriate amounts of Pt-C powder, ionomer, and deionized water were mixed by the rotating and revolving mixer at a rotation speed of 2000 rpm and revolution speed of 800 rpm for 15 min. After that, a certain amount of ethanol was added to the aqueous slurry, and the resulting mixture was mixed under the same conditions. The composition of the final slurry obtained by the two-step method was identical to that of the slurry prepared by the one-step method, as shown in Table 1 (see Table 2, procedure 2). Both slurries were characterized by several analytical techniques and then used to fabricate PEFC electrodes to evaluate the performances of their corresponding fuel cells.

Furthermore, to determine the effects of the particle concentration and solvent type on the amount of adsorbed ionomer, slurry with a different composition was prepared

Fig. 1. Slurry preparation procedures.

Table 1. Slurry composition for the anode

Ethanol conc. (mass %)	0	7.5	15	22.5	30
Pt/C (g)			2.11		
Nafion (g)			12.9		
Water (g)	11.5	9.79	8.07	6.35	4.63
Ethanol (g)	0	1.72	3.43	5.16	6.83
Pt/C conc. (mass %)			8.0		

Table 2. Slurry composition for the two-step method

	Ethanol conc. (mass %)	7.5	15	22.5	30
Procedure 1	Pt/C (g)		2.11		
	Nafion (g)		12.9		
	Water (g)	9.79	8.07	6.35	4.63
	Pt/C conc. (mass %)	8.5	9.1	9.9	11
Procedure 2	Ethanol (g)	1.72	3.43	5.16	6.83
	Pt/C conc. (mass %)		8.0		

Table 3. Slurry composition for the discussion of the dilution effect on the adsorbed ionomer amount

Ethanol conc. (mass %)		7.5	15	22.5	30
Procedure 1	Pt/C (g)			2.11	
	Nafion (g)			12.9	
	Water (g)	7.81	4.52	1.58	0.685
	Ethanol (g)	1.56	2.81	3.78	4.03
	Pt/C conc. (mass %)	8.7	9.5	10	11
Procedure 2	Water (g)	1.98	3.56	4.78	5.10
	Ethanol (g)	0.151	0.627	1.38	1.69
	Pt/C conc. (mass %)			8.0	

as well. First, the slurry with a particle concentration greater than that of the slurry listed in Table 1 (above 8 mass %) was obtained using the same solvent mixture with an ethanol concentration of 7.5, 15, 22.5, or 30 mass % (see **Table 3**, procedure 1). The approximate volume of the solvent mixture in this procedure was maintained constant. After that, the slurry prepared in procedure 1 was diluted by the water-ethanol mixture with the same ethanol concentration to obtain the slurry with the composition specified in Table 1.

2.3 Slurry characterization

The slurry prepared in section 2.2 was characterized as follows. First, its packing ability was evaluated by conducting a centrifugal sedimentation test. In this test, the slurry was transferred to a test tube to achieve the initial height of 9.0 cm and then centrifuged at a centrifugal acceleration of 180 G for 20 days. The time change of the sediment height was periodically checked, and when no height changes were observed after 1 d of centrifugation, the resulting height was considered the final height at the equilibrium state. The final packing fraction, Φ [–], was determined from the measured final height of the sediment, H [m], using the following material balance equation:

$$\Phi = \frac{h}{H} \phi \quad (1)$$

Here, h [m] is the slurry height in the test tube ($h = 0.09$ m), and ϕ [–] is the volumetric fraction of the particles in the initial slurry.

In addition, the amount of adsorbed ionomer was determined. Clear supernatant was obtained by the centrifugation of the prepared slurry at a centrifugal acceleration of 1060 G for 1 day. The sampled supernatant was analyzed by a thermogravimetric analyzer (TGA-50, Shimadzu Corp., Japan) to determine the concentration of non-adsorbed ionomer. By subtracting the amount of non-adsorbed ionomer from that of added ionomer, the amount of ionomer adsorbed onto Pt-C powder was calculated. During thermogravimetric analysis, around 90 mg of the sampled supernatant was placed inside the instrument, and the decrease in the sample weight was measured while increasing the specimen temperature at a rate of 15 °C/min. The weight loss in the temperature range from 350 to 500 °C corresponded to the amount of non-adsorbed ionomer, as shown in **Fig. 2**.

Fig. 2. A typical thermal analysis curve obtained for the ionomer solution.**Table 4.** Viscosity and refractive index of the mixed solvent

Ethanol conc. (mass %)	0	7.5	15	22.5	30
Viscosity (mPa·s)	1.00	1.10	1.57	1.90	2.70
Refractive index (—)	1.33	1.34	1.34	1.35	1.35

The size distribution of ionomer in the solvent was measured as follows. A solution with the same composition except for Pt-C powder was prepared first (see Table 1). For the one-step method, the solution was obtained by mixing certain amounts of ionomer, deionized water, and ethanol at a rotation speed of 2000 rpm and revolution speed of 800 rpm for 15 min. In the two-step method, the ionomer was mixed with deionized water at a rotation speed of 2000 rpm and revolution speed of 800 rpm for 15 min, after which a specified amount of ethanol was added to the resulting mixture followed by mixing under the same conditions. The prepared solution was analyzed by dynamic light scattering (DLS; LB-550, Horiba, Ltd., Japan) to determine the ionomer size distribution. The viscosities and refractive indexes of the mixed solvent are listed in **Table 4**.³⁵⁾ The refractive index of Nafion was chosen 1.38 according to the previous report.³⁶⁾

2.4 Electrode fabrication and evaluation of the cell performance

It is well known that the oxygen transport at the cathode often worsens the PEFC performance;^{37),38)} hence, cathodes in this work were prepared under different conditions. Cathodes for the fuel cell were fabricated by a doctor blade method from the slurries obtained by the one-step and two-step methods. First, the slurry was coated onto a polytetrafluoroethylene sheet. The blade gap was controlled properly to maintain a constant weight per unit area of Pt (0.35 mg/cm²). The casting rate of the sheet was 0.60 m/min. The cast catalyst layer was dried at 25 °C for about 1 h.

The anode was fabricated from Pt-GC slurry (Tanaka Kikinzoku Kogyo, Japan) and ionomer (D2020, Dupont, USA) by the same doctor blade method. The weight of the anode per unit area of Pt was 0.05 mg/cm². The conditions

for the slurry preparation and doctor blade casting steps were the same during anode fabrication.

A membrane electrode assembly (MEA) was produced by the hot pressing of the cathode, anode, and electrolyte film (NR211, Dupont, USA) at a temperature of 150°C, applied pressure of 0.8 MPa, and holding time of 10 min. During the installation of the MEA into the test cell, carbon paper (SGL29BC, SGL Carbon Japan Co., Ltd., Japan) was attached to both sides of the electrodes.

To record I - V curves for all MEAs prepared from various slurries, the electric voltage was measured while changing the electric current. Here, each value of the electric current was kept constant for 300 s to stabilize the measured electric voltage. The I - V curve was obtained using the current density determined by dividing the electric current by the electrode area of 10 cm². To measure the electric voltage, H₂ and O₂ gases were supplied to the anode and cathode at gas rates of 6.7×10^{-5} and 2.4×10^{-4} , respectively. The gas pressure, relative humidity, and cell temperature were 200 kPa, 40%, and 80°C, respectively.

Generally, PEFC exhibits a typical polarization breakdown with increasing current density due to the activation (decrease in the reaction rate), ohmic (resistance), and concentration (gas transport) losses.³⁷⁾ In all three cases, the reaction rate normally depends on the surface area of Pt in the catalyst layer. In this study, we used only one type of Pt-C powder; thus, it can be assumed that the decrease in the reaction rate is hardly affected by the slurry preparation conditions. Therefore, the resistance and gas transport losses were dominated by the transport resistances of protons and oxygen species, thus, the transport resistances of protons, R_{ion} , and oxygen, R_{other} , was determined by using the following electrochemical diagnostic method.

From the analysis of the Nyquist diagram obtained by measuring the electrochemical impedance, the transport resistance of protons, R_{ion} , in the catalyst layer was determined.³⁹⁾ The evaluation device consisted of a potentiogalvanostat, a frequency analyzer, and the fabricated MEA. The measurement was conducted at a relative humidity of 40%, cell temperature of 80°C, gas flow rate of 8.3×10^{-6} Nm³/s, and pressure of 100 kPa. The impedance was determined by processing the signal corresponding to the modulation potential applied to the MEA.⁴⁰⁾

In addition, by conducting limiting current measurements, the total oxygen transport resistance in both the gas diffusion and catalyst layers, R_{total} , can be evaluated and used to extract the gas transport resistance of the catalyst layer, R_{other} .^{27),28)} The limiting current was measured at a relative humidity of 90%, cell temperature of 80°C, hydrogen flow rate of 6.7×10^{-5} Nm³/s, and oxygen flow rate of 2.5×10^{-4} Nm³/s. The current was measured by varying the gas pressure between 125, 154, and 202 kPa, and oxygen concentration between 1.0, 1.3, 1.8, and 2.6 mass %. The gas transport resistance of the catalyst layer, R_{other} , was determined from the plot of R_{total} , where the intercept of the plot expresses R_{other} .^{27),28)}

The detailed methods to determine R_{ion} and R_{other} were described in our previous paper.³⁴⁾

3. Results

3.1 Slurry characterization

Figure 3 shows the final packing fractions of the slurries prepared by the one-step and two-step methods after centrifugal sedimentation. The experimental error for measuring the packing fraction was less than 0.01 when checking it by using the slurry with an ethanol concentration of 7.5 mass %. Their values increased with an increase in the ethanol concentration as reported in our previous study;³⁴⁾ however, the difference between these two slurries was not significant except for the slurry with an ethanol concentration of 7.5 mass %.

Figure 4 shows the ionomer size distribution measured by DLS while changing the ethanol concentration for both the one-step and two-step methods. From this result, the size of ionomer became more than 1 μm in most slurries. This is probably due to aggregation of the ionomer as shown in previous studies in which the ionomer size was measured not only by DLS^{41),42)} but also Cryo-TEM observation.⁴³⁾ In the first method, the mean size of ionomer species decreased with an increase in the ethanol concentration as shown in Fig. 5. This trend is likely due to the different dispersion states of ionomer particles caused by their different affinities to the solvent at various ethanol concentrations.^{21),44)} Furthermore, the mean ionomer size in the two-step method was almost identical to that in the one-step method when the mixed solvents had the same final ethanol concentrations.

Figure 6 shows the amount of adsorbed ionomer onto Pt-C powder determined at various ethanol concentrations. For the one-step method, its magnitude decreased with an increase in the ethanol content, which was consistent with results of our previous study.³⁴⁾ This trend can be explained by the higher degree of hydrophilicity of the Pt-C particle surface caused by ethanol adsorption,^{45),46)} which resulted in the decrease in the amount of ionomer adsorbed via hydrophobic interactions.^{37),47),48)} For the two-step method, the amount of adsorbed ionomer also decreased as the ethanol concentration increased; however, its value was larger than the magnitude obtained for the

Fig. 3. Packing fractions of the sediments for gravitational sedimentation testing.

Fig. 4. Size distribution of the non-adsorbed ionomer measured by DLS.

Fig. 5. Effect of the ethanol concentration in solvent on the median size of ionomer.

Fig. 6. Effect of the ethanol concentration in solvent on the adsorbed amount of ionomer.

one-step method despite the same final compositions of the corresponding slurries.

3.2 Cell performance evaluation

Figure 7 shows the I - V curves recorded for the MEAs prepared from different slurries by the one-step and two-

step methods. Except for the ethanol concentration of 7.5 mass %, the curve obtained in the second method was better than that obtained in the first method. Figures 8 and 9 show the transportation resistances of protons, R_{ion} , and oxygens, R_{other} , respectively. For the oxygen transportation resistance, the R_{other} values obtained in the two-step method was smaller than those determined in the one-step method at all ethanol concentrations. For the proton transportation resistance, the R_{ion} magnitudes obtained in the two-step method were also smaller than those measured in the one-step method at all ethanol concentrations except for 7.5 mass %. The reason for this phenomenon remains unclear; however, from these results, it can be concluded that the performance of the cell fabricated by the two-step method was better than that of the cell produced by the one-step method.

4. Discussion

In this work, possible reasons for the improvement of the cell performance observed for the two-step method despite the identical final composition of the slurry were discussed. First, the particle packing ability of each slurry did not significantly changed except for the slurry with an ethanol concentration of 7.5 mass %, as shown in Fig. 3. This is probably because the slurry with an ethanol concentration of 7.5 mass % showed the great difference of the adsorbed amount of ionomer between one-step and two-step method, that is, the relatively large amount of adsorbed ionomer caused aggregation of Pt-C, resulting in the lower packing fraction for two-step method. However, the packing fraction difference was less than 0.01 and the MEAs were fabricated by the hot pressing of the catalyst layers at 0.8 MPa, suggesting that the effect of the particle assembly catalyst layer (including the particles packing fraction and void structure) should not be remarkable. Hence, we focused on the amount of adsorbed ionomer and size of non-adsorbed ionomer species as the main factors affecting the cell performance.

In our previous work,³⁴⁾ it was demonstrated that the proton transportation resistance decreased as the amount of ionomer adsorbed onto Pt-C powder increased when

Fig. 7. I - V curves recorded at various slurry preparation conditions. Fig. 8. Effect of the ethanol concentration in solvent on R_{ion} . Fig. 9. Effect of the ethanol concentration in solvent on R_{other} .

the added amount of ionomer was constant. These results indicate that the presence of ionomer around Pt-C powder formed a network structure that facilitated proton transport. As shown in Fig. 6, the proton transportation resistance decreased after the amount of adsorbed ionomer increased in both methods at the same slurry composition. **Figure 10** displays the relationship between the amount of adsorbed ionomer and proton transportation resistance. It shows that the latter parameter decreases with ionomer adsorption, which was in good agreement with the results of our previous work.³⁴⁾

Possible reasons for the high amount of adsorbed ionomer in the two-step method as compared with that

obtained for the one-step method include (1) mixing Pt-C powder and ionomer at a higher particle concentration and/or (2) mixing them in pure water during procedure 1 of the two-step method. It was reported previously that polyelectrolytes adsorbed onto alumina particles surface very strongly and that desorption did not occur after dilution, although the studied system was not a PEFC slurry.^{49),50)} It was also reported that the amount of adsorbed polyelectrolyte increased as the particle concentration increased when the added polyelectrolyte amount per unit particle surface area was constant.⁵⁰⁾ This phenomenon has not been studied in detail yet; thus, both mechanisms likely affected the increase in the adsorbed amount of ionomer.

Fig. 10. Effect of the adsorbed amount of ionomer on R_{ion} .

Fig. 11. Effect of dilution on the adsorbed amount of ionomer.

To verify this hypothesis, the adsorbed amount of ionomer was measured for the slurry prepared at a higher particle concentration, after which the slurry with the composition listed in Table 1 was obtained by dilution using a mixed solvent with the same ethanol concentration (see Table 3). The amount of adsorbed ionomer determined for this system is presented in Fig. 11. It shows that in contrast to the slurry prepared by the two-step method, the adsorbed ionomer amount does not increase in the slurry obtained by the dilution method. These results indicate that the increase in the adsorbed ionomer amount observed for the two-step method should be mainly attributed to the preparation of aqueous slurry in the first step. In water, ionomer species strongly adsorb onto Pt-C powder due to hydrophobic interactions, which decrease the amount of desorbed ionomer due to dilution. For this reason, the adsorbed ionomer amount in the two-step method was larger than that in the one-step method.

Meanwhile, it was also shown that the oxygen transportation resistance of the PEFC catalyst layer decreased as the size of non-adsorbed ionomer species decreased.³⁷⁾ As shown in Fig. 4, the size difference between the non-adsorbed ionomers in the one-step and two-step methods is relatively small as compared with the difference between the adsorbed ionomer amounts, although the non-adsorbed

Fig. 12. Effect of the mean diameter of non-adsorbed ionomer on R_{other} .

ionomer size itself decreased as the ethanol concentration increased. Figure 12 displays the relationship between the oxygen transportation resistance R_{other} and the mean size of non-adsorbed ionomer. It shows that the R_{other} value strongly depends on the size of non-adsorbed ionomer species and decreases with a decrease in the mean ionomer size; however, its difference between the one-step and two-step methods is not significant. From these results, it can be concluded that not only the mean ionomer size but also the ionomer state around Pt-C powder affected the decrease in R_{other} . The larger amount of adsorbed ionomer in the two-step method at a same slurry composition resulted in its more homogeneous distribution in the catalyst layer after drying. The formation of large ionomer aggregates adsorbed on the powder surface when water was used as the solvent was reported in previous works.^{21),34),41)} When the ionomer state remained the same, its thick adsorbed layer disturbed the oxygen transportation to increase R_{other} . However, in the two-step method, the addition of ethanol after preparing the aqueous slurry changed not only the size of non-adsorbed ionomer particles, but also the parameters of adsorbed ionomer species (for example, the thickness of their layer). Owing to the change in the state of adsorbed ionomer, the oxygen transportation resistance decreased. In contrast, in the one-step method, the size of non-adsorbed ionomer species was also small at relatively high ethanol concentrations; however, their amount exceeded that obtained for the two-step method. Such a large number of non-adsorbed ionomer particles may move and plug the interparticle micropores during drying and thus increase the transportation resistance of oxygen. Hence, the oxygen transportation resistance in the one-step method was larger than that in the two-step method although the mean size of non-adsorbed ionomer particles was hardly changed by the slurry preparation procedure.

Figure 13 shows the SEM cross-sectional images of the catalyst layers fabricated by the one-step and two-step methods. First, the packing state of Pt-C powder hardly changed among the all catalyst layers used in this work. In both the one step and two step methods, the boundaries of

Fig. 13. Microstructure of the catalyst layer cross-section observed by SEM. (a) 1 step method and (b) 2 step method.

Pt-C particles become clearer as the ethanol concentration increases because the adsorbed ionomer amount decreases. The images obtained in the middle ethanol concentration range of 7.5–22.5% display the powder obtained by the two-step method more clearly. These data qualitatively match the results of centrifugal sedimentation testing and measurements of adsorbed ionomer amounts reported above.

The microstructure of the catalyst layer fabricated by the two-step method is schematically drawn in **Fig. 14**. Because the aqueous slurry was prepared first, the amount of adsorbed ionomer increased. Moreover, by adding ethanol into this slurry, the thickness of the ionomer layer and mean size of non-adsorbed ionomer particles de-

creased. In both processes, ionomer species are attached homogeneously to the Pt-C particles in the catalyst layer after drying, simultaneously decreasing the transportation resistances of protons and oxygens. Therefore, the two-step method can potentially improve the performance of PEFCs without adding more Pt and ionomer species and thus reduce their production cost.

5. Conclusions

In order to lower the transportation resistances of protons and oxygens in PEFCs, the effects of adsorbed ionomer amount and mean size of non-adsorbed ionomer species on the slurry characteristics and cell performance were investigated. To increase the adsorbed amount of

Fig. 14. Schematic illustration of the microstructure and transport resistances R_{ion} and R_{other} .

ionomer onto Pt-C powder and decrease the mean size of non-adsorbed ionomer, a two-step method, in which electrode slurry was prepared in water first followed by ethanol addition, was examined. By determining the relationship between various slurry parameters such as the particle packing ability, adsorbed ionomer amount, mean size of ionomer particles, and cell performance characteristics such as the I - V curve, the transportation resistances of protons and oxygen species were obtained, and the following conclusions were drawn.

(1) The adsorbed ionomer amount increased after changing the slurry preparation process from the one-step to the two-step method although the final slurry had the same composition.

(2) The increase in the adsorbed amount of ionomer observed for the two-step method should be attributed to mixing Pt-C powder and ionomer in water.

(3) The transportation resistances of protons and oxy-

gens can be reduced by the slurry preparation via the two-step method without adding more Pt catalyst or ionomer species.

From these conclusions, a potential way to improve the PEFC cell performance from a processing viewpoint can be proposed.

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