



Dependence of polymer electrolyte fuel cell performance on preparation conditions of slurry for catalyst layers

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ABSTRACT

The catalyst slurry used to form the catalyst layer of a polymer electrolyte fuel cell (PEFC) must be mixed for a sufficiently long period for good and stable cell performance. However, the optimum mixing duration must be determined from the viewpoint of process design. We prepared slurries with various amounts of Nafion and examined the influence of the slurry mixing time on the viscosity of the slurry, the size of the pores and distribution of elements in the catalyst layer, and the cell voltage of the PEFC. We found that when the Nafion content is optimum, these properties change gradually and stabilize after a sufficiently long mixing time as the progression of the adsorption of Nafion to catalyst particles. However, when the Nafion is comparatively low, although the properties stabilize after sufficient mixing, the Nafion molecules are first dispersed and then localized around aggregates. This reduces cell performance when mixing excessively long. These differences in the structural and electrochemical behaviors could be predicted by measuring the variation in viscosity during the mixing process.

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1. Introduction

Polymer electrolyte fuel cells (PEFCs) are seen as promising, yet environmentally friendly, alternatives to the rapidly depleting fossil fuel resources. But the manufacture of PEFCs with practical performance levels and production costs will require the use of various new materials, most of which are still in development. The key part of a PEFC is its membrane electrode assembly (MEA), which consists of an ion-exchange membrane and catalyst and diffusion layers. Of these, the catalyst layer, which generally comprises expensive Pt particles that catalyze the redox reactions of hydrogen and oxygen, contributes significantly to the price of current PEFCs. Thus, various researchers have attempted to develop modified Pt-type or Pt-alloy-based catalysts and optimize the size and shape of the catalyst particles [1].

In addition to the redox reaction, the gas, proton, and electron transports through the catalyst layer are significant determiners of fuel cell performance. In general, PEFC catalyst layers are produced by coating and drying a slurry comprising a mixture of Pt-loaded carbon black (Pt/C) particles and a solution of the polymer electrolyte (Nafion). Electrons and protons are conducted via the percolation of the carbon black particles and Nafion molecules,

respectively. The gaseous fuels (hydrogen and oxygen) and waste product (water) pass through the pores of the dried slurry. Gode et al. [2] pointed out that the pore structure has a significant influence on cell performance. Therefore, we must establish a method to simultaneously control both the internal pore structure and percolation.

Evaluation and control of the pore structure can be conducted in various ways. The catalyst layer will have pores of various sizes, corresponding to catalyst particles, carbon black particles, their aggregates, etc. Scanning electron microscopy is the most common methodology used to observe the pore structure. Pore size and volume can be measured by gas adsorption [2–4] or mercury intrusion [2,5–8], depending on the pore size of interest. The influence of the Nafion content on the pore structure and cell performance has been investigated by many groups. A Nafion-rich slurry, on the one hand, produces a less-porous catalyst layer and inhibits the percolation of carbon black and gas diffusion. A Nafion-poor slurry, on the other hand, will have insufficient Nafion molecules for proton conduction. Thus, the Nafion content must be optimum. This has been reported to vary from 25 to 40 wt%, depending on the composition of the slurry [2,5,9]. In addition, a pore-forming additive can be used to produce pores of the desirable size in the catalyst layer and thereby enhance the transport of air or oxygen [3,10]. Our group has been investigating the effect of the coating process and the slurry composition on the structure of the catalyst layer. High-speed coating has been found to improve the performance of the catalyst layer when

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using a slurry of appropriate viscosity [11]. The effect of the coating speed cannot be observed clearly as reducing slurry viscosity, but an extremely low viscosity causes a drastic decrease in performance and reproducibility independent of coating speed owing to instability of wet slurry film [12]. As a result, we found that the optimum composition of the slurry, at which we can neglect the effect of the coating speed on the cell performance, shows preferable agreement with that proposed by various researchers.

In most studies conducted thus far, including ours, slurries were prepared by mixing and ultrasonication. These studies, however, did not have clearly defined mixing durations and sensuous criterion, such as viscous, homogeneous, and smooth, were often used to terminate preparation process of slurry. Adsorbed structure of Nafion molecules on Pt/C particles forms in slurry preparation process and this will affect the structure of the catalyst layer and, therefore, the performance of the PEFC cell. However, it remains unclear as to what changes actually occur in the internal structure during the preparation process and what criteria can be used to identify the optimum conditions for slurry preparation. We must have no reasonable confidence in slurry preparation process when we coated it to make a catalyst layer.

In this work, we investigated the influence of the mixing time on the properties of the slurry and resultant catalyst layer and on the performance of the resultant PEFC. The slurry was characterized by its viscosity, which is significantly affected by particle aggregation or the formation of internal structures. The pore structure and distribution of elements in the dried catalyst layer were measured. Further, the mechanism of dispersion and aggregation of Pt/C particles and Nafion molecules in the slurry is discussed.

2. Experimental

2.1. Materials

The slurry was made from Pt/C nanoparticles (HiSPEC 4000 fuel cell catalyst, Johnson Matthey Plc.), a 5 wt% Nafion perfluorinated resin solution (Sigma–Aldrich Inc.), and isopropanol (IPA). The Pt/C nanoparticles were made from carbon powder (Vulcan XC-72, Cabot Corp.) on which smaller platinum particles of 2–3 nm in diameter are loaded to a concentration of 40 wt%. We prepared slurries of the two different compositions listed in Table 1. This table also shows the volume fraction of Pt/C particles, which is denoted by ϕ , and the Nafion content (weight concentration of Nafion in a dried catalyst layer). The volume of Pt/C particle was estimated from the specific densities and weight fractions of carbon and platinum. The same dispersion medium, comprising a mixture of Nafion solution and IPA, is used for both slurries. Thus, the difference between the two slurries lies solely in the volume fraction of suspended particles (Pt/C) or the Nafion content. Since the optimum Nafion content has been reported to lie between 25 and 45 wt%, a Nafion content of 33 wt% was assumed to be optimum, and therefore, a Nafion content of 20 wt% was considered insufficient in relation to the Pt/C content.

2.2. Preparation of slurry and viscosity measurement

The materials, in the appropriate proportions (Table 1), were mixed in a glass bottle (40 mm i.d.) purged with nitrogen gas, because the mixture of IPA and Pt catalyst is flammable in air. The volume of the mixture and liquid height were 20 ml and 16 mm, respectively. This was mixed for 0.5–24 h with a 25-mm-long magnetic stirrer bar at 500 rpm. Ultrasonication was not employed.

The slurry viscosity was measured at 25 °C using a stress-controlled rheometer (SR-5, Rheometric Scientific Inc.) with a cone-plate geometry (cone diameter, 4 cm; angle, 0.04 rad). To prevent a change in slurry composition by the evaporation of IPA during the measurement, the cone-plate fixture was covered by a plastic box filled with IPA vapor. However, the reduction in slurry volume by the amount used for the viscosity measurement will influence the mixing efficiency. Thus, we prepared several batches of slurry and used one bottle of slurry for two measurements at most. Therefore, despite the slurry composition being the same over the various mixing times on the plot, the batch of slurry itself is not the same.

2.3. Production and evaluation of catalyst layer

Teflon sheets were coated with the slurry by the doctor blade method with a coating gap of 100 μm and coating speed of 100 mm s^{-1} . The coating experiments were carried out by a speed-controlled coater (CC-300, Imoto Machinery Co., Ltd.) and a gap-controlled film applicator (SA-204, Tester Sangyo Co., Ltd.).

If the slurry is allowed to dry in the air at room temperature, the surface and internal structure of the dried film will be difficult to control because they are influenced by unpredictable factors such as the room temperature, relative humidity, and air flow. Thus, we used a pressure-controlled drying technique [13] that avoids crack formation by using reduced drying rate. The slurry-coated sheet was placed with a drying solvent in a dessicator held at a pressure slightly higher than the saturation vapor pressure of IPA (hold pressure) by using a pressure control system. After a fixed time (hold time), the pressure was reduced again in order to remove as much of the dispersion medium as possible. In this study, the pressure of the dessicator was reduced from atmospheric pressure to 57 hPa in 3 min and held for 1 h. Then, the pressure was reduced further to less than 20 hPa.

The dried slurry obtained as above was used to form a MEA by the decal method [14] to allow evaluation of the cell performance. The dried catalyst layers were hot-pressed on to both sides of a Nafion 112 membrane for 5 min with the pressure of 100 kgf cm^{-2} . The Teflon sheets were then removed. Carbon cloth was used as the gas diffusion layer and current collector on both sides of the membrane to form the MEA. Thus, a MEA of size 5 cm \times 5 cm was installed with the carbon cloth in a single test cell. The weight of catalyst layer was calculated by subtracting the weight of removed Teflon sheet from that of the sheet with the catalyst coat. Neat hydrogen and air were supplied, with coefficients of utilization of 70% for hydrogen and 40% for air. Hydrogen was humidified at 80 °C and air at 70 °C. After aging at a current density of 0.3 A cm^{-2} for more than

Table 1
Composition of slurry used for production of catalyst layer.

Weight fraction (wt%)			Volume fraction of Pt/C, ϕ	Nafion content (wt%)	
Pt/C	Nafion solution	IPA		Dispersion medium of slurry	Dried slurry
6.8	67.8	25.4	0.018	3.63	33.3
12.7	63.5	23.8	0.035		20.0

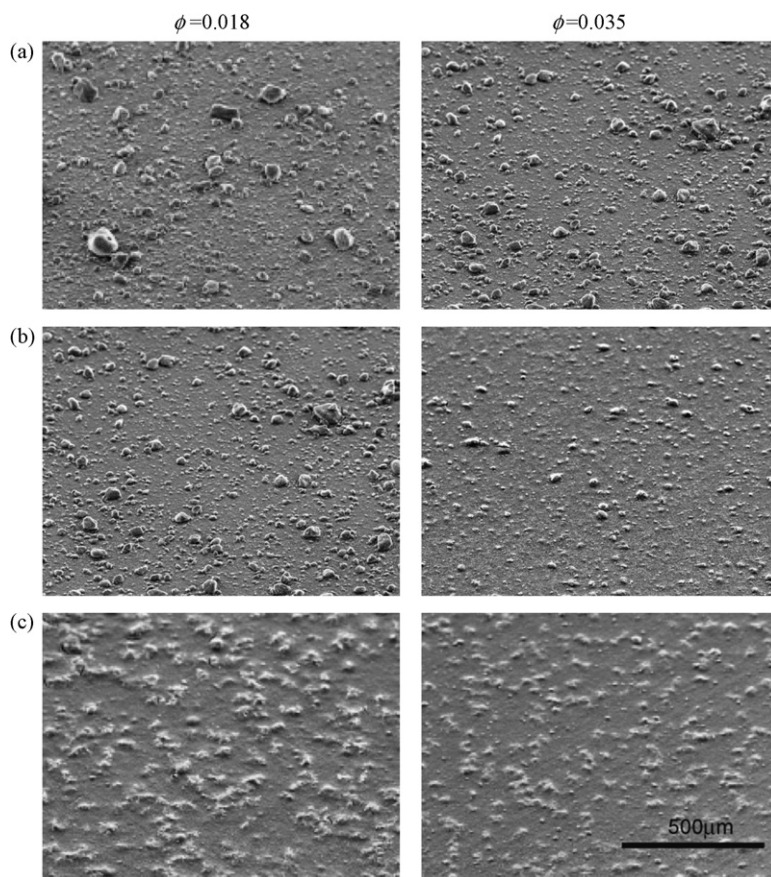


Fig. 1. SEM images of surface of catalyst layer produced from slurries mixed for: (a) 0.5 h, (b) 4 h and (c) 24 h.

8 h, the cell voltages were measured at various output current using a single-cell testing apparatus (FC5400, Chino Corp.). The internal resistance of the cell was also measured at the same time. The sum of the cell voltage and the voltage drop due to the internal resistance is indicated as the “iR-free” cell voltage. The influence of the anode structure on cell performance could be ignored [11] because of the rapid kinetics of hydrogen oxidation compared with the rate of oxygen reduction at the cathode. We then used catalyst layers produced from slurries mixed for different periods of time at the cathode, whereas the slurry used for the anode catalyst was kept the same. In addition, both electrodes were produced using slurry of the same composition.

The pore size distribution was measured by a mercury porosimeter (Auto Pore 9500, Shimadzu Corp.). And a scanning electron microscope (SEM; JSM-5610LVS, JEOL Co Ltd) was used to observe

the surface of the catalyst layer and also to conduct elementary analysis with an energy-dispersive X-ray spectrometer (EDS).

3. Results and discussion

3.1. Change in slurry properties during mixing process

Just after the addition of the Pt/C particles to the Nafion solution, the particles stick together to form clusters, in which particles will be packed tightly. Thus, the voids in these clusters should be negligibly small, allowing each cluster to be treated as a large particle. As the mixing proceeded, the particles are first dispersed and then tend to stick together again to form loosely packed aggregates. The transition of particles from clusters to aggregate must significantly affect the structure after drying. Fig. 1 shows SEM micrographs of

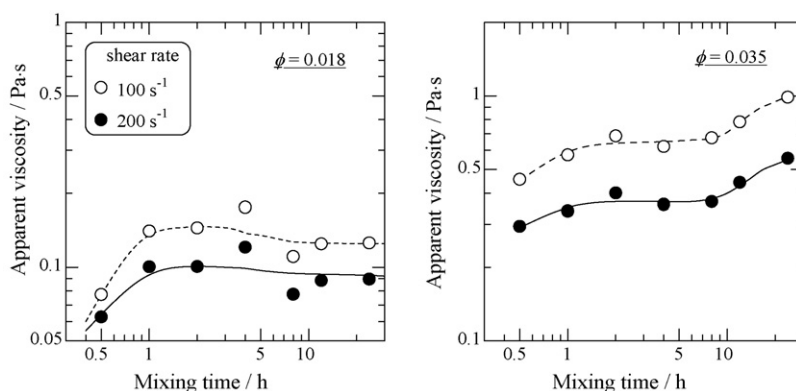


Fig. 2. Variation in slurry viscosity with mixing time.

the surfaces of the dried slurries mixed for various durations. At a short mixing time, clusters were observed on the surface of a catalyst layer. At 4 h, the size of clusters reduced remarkably and we could scarcely observe fractures. With further mixing, many aggregates, which are smaller than clusters, were observed on the surface of a catalyst layer.

The configuration of the clusters is much clearer than that of the aggregates in Fig. 1. This suggests that the particles in a cluster are tightly bound and any voids inside clusters will be much smaller than those in an aggregate. And furthermore, the total volume of aggregates will be larger than that of individual particles. As a result, the large apparent volume fraction of aggregates will increase slurry viscosity. When the dispersal of particles reaches a steady state during the mixing process, the slurry viscosity remains constant. Thus, variations in the slurry viscosity reflect changes in the dispersal and aggregation of the particles. In addition, the dispersed particles must be bonded with the polymer in the polymer solution. Thus, the interaction between particle and polymer also affects slurry viscosity.

The apparent viscosities of both types of slurries are plotted in Fig. 2 as a function of mixing time at shear rates of 100 and 200 s⁻¹. For the slurry with $\phi = 0.018$, the viscosity increased quickly just after the start of mixing and reached a constant value corresponding to the shear rate applied. However, for the slurry with $\phi = 0.035$, the viscosity increased gradually during the 24 h of mixing, with a small drop between 2 and 4 h. These different results suggested that the dispersibility of the particles in the polymer solution is significantly influenced by the number ratio of the suspended particles to the bounded polymer molecules. Further, the results suggest that the optimum mixing conditions must differ with the slurry composition.

3.2. Structure of catalyst layer

After 24 h of mixing, the aggregates in the slurry with $\phi = 0.018$ were significantly smaller than those in the slurry with $\phi = 0.035$ (Fig. 1). The sizes of the aggregates correspond with the measured viscosities. In other words, in a slurry with a low number density of particles, the suspended particles are well separated from each other, making the dispersed state more stable. However, particles form large aggregate in a slightly concentrated slurry. Therefore, the structure of the aggregates in the slurry as well as in the dried

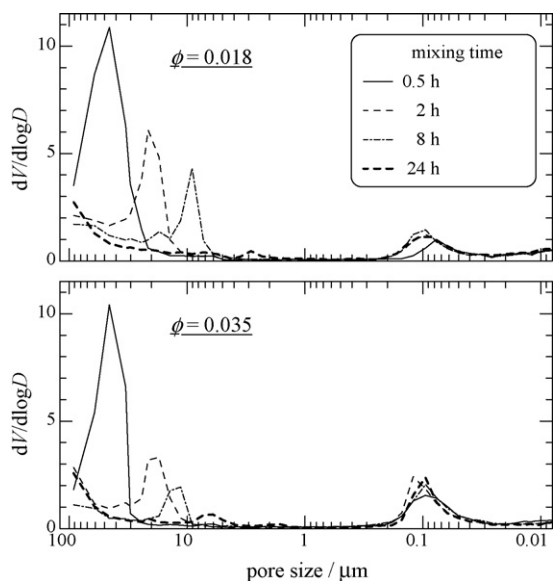


Fig. 3. Pore size distribution of catalyst layer.

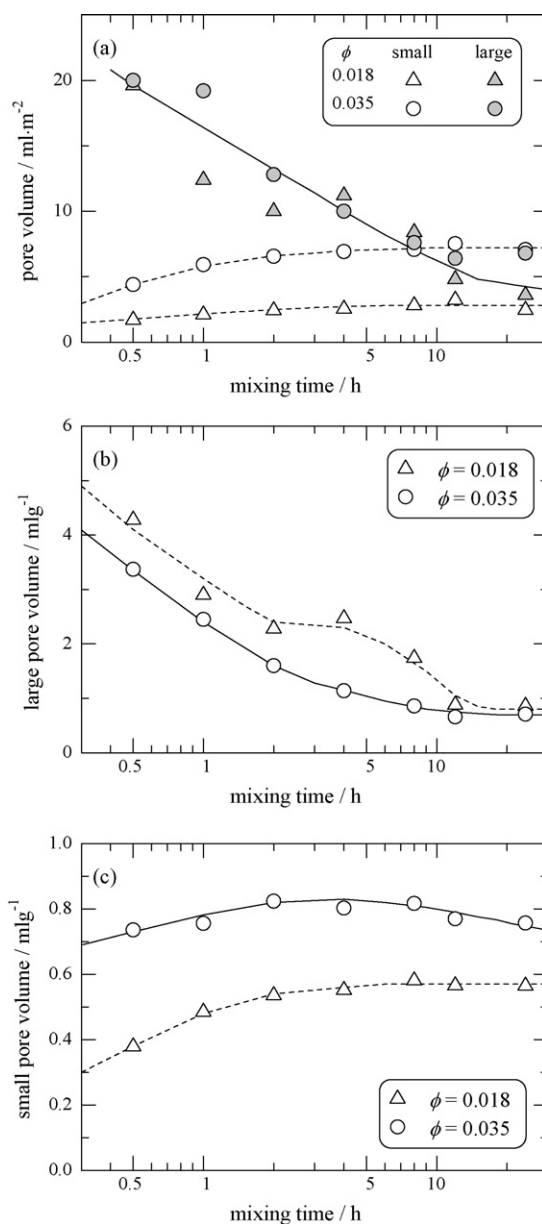


Fig. 4. Variation in pore volume with mixing time: (a) total pore volume, (b) specific large pore volume, and (c) specific small pore volume.

catalyst layer must change according to the composition and mixing time. Furthermore, it is easily conceivable that the internal structure of clusters or aggregates in the slurry affects the internal structure of the dried catalyst layer.

To better understand the internal structure of the catalyst layer, the pore size distribution of the catalyst layer was measured by mercury porosimetry; the results are shown in Fig. 3. We found that the catalyst layer had roughly two types of pores with different size ranges. Watanabe et al. [15] reported that both small and large pores can be found, and that they corresponded to the spaces within aggregates of Pt/C particles and the spaces between such individual aggregates, respectively. We found large pores with sizes in the range from 1 to 100 μm , depending on the mixing time. Additionally, cracks of size 10 or 100 μm were observed on the surface of the catalyst layer in SEM images, and their total volume was comparatively large, especially at initial stages of mixing. Thus, large pores included both cracks and the spaces between aggregates. The sizes of large pore reduced significantly as mixing progressed. For both

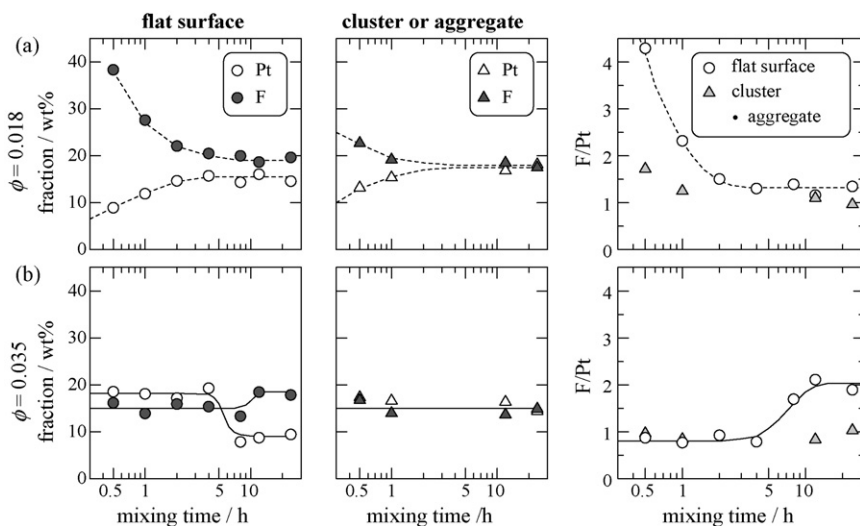


Fig. 5. Weight fractions of F and Pt on the surface of catalyst layer.

slurries, in contrast to the change in the viscosity, the sizes of large pores remained almost the same with the mixing time. The small pores were approximately $0.1 \mu\text{m}$ in size and also did not change in size with mixing. Thus, the small pores represent voids between aggregates, as reported by Watanabe et al., because the sizes of the pores in the aggregates must be on the order of that of the Pt/C particles, i.e., several tens of nanometers. Furthermore, there is no clear dependence of the void size on the composition and mixing time.

For a more quantitative discussion of the pore size distribution, the volumes of the large and small pores were plotted against the mixing time (Fig. 4). The large pore volume was calculated as the cumulative volume of pores from 1 to $60 \mu\text{m}$ in size, and the small pore volume, the cumulative volume of pores from 0.03 to $0.35 \mu\text{m}$ in size. The pore volume normalized by the area of the catalyst layer, $5 \text{ cm} \times 5 \text{ cm}$, was shown in Fig. 4(a). It was found that the volume of large pores changed with almost the same trend, independent of the composition until 12 h of mixing, whereas the small pore volume showed a definite dependency on composition. The results indicated that the change in surface structure influenced the pore volume. From SEM images (Fig. 1), cracks drastically decreased as mixing progressed. Therefore, because the volume of cracks is much larger than that of space between aggregates, the decrease in large pore volume can be primarily attributed to the decrease in the volume of cracks. After 24 h of mixing, catalyst layers made from both types of slurries had the same specific volume of large pores (Fig. 4(b)) and distinguishable fractures could not be observed on the surface. These results suggested that crack formation was considerably suppressed and that the large pores were mainly of the spaces between aggregates, which are roughly distributed over the whole of the catalyst layer.

In contrast, the volume of small pores (void) increased with the mixing time until 4 h (Fig. 4(c)). Further mixing cause a small decrease in void volume for the slurry with $\phi = 0.035$, whereas the void volume remained constant in the catalyst layers made from the slurry with $\phi = 0.018$. The initial increase in void volume is caused by the breaking up of clusters, which have small voids, and the formation of loosely packed aggregates of Pt/C particles and Nafion molecules. Consequently, the slight decrease in void volume for the slurry with $\phi = 0.035$ suggests that the interaction between the Nafion molecules and Pt/C particles changes when the slurry is mixed for too long. From the above, we can conclude that for a concentrated slurry, excessive mixing will decrease the number of

cracks on the surface of the dried catalyst layer, but will not stabilize the void structure.

3.3. Elementary analysis of surface of catalyst layer

In a Nafion solution, Nafion molecules form aggregates was reported to be several hundred nanometers in size [16]. Similarly, Pt/C particles also form aggregates in the dispersion medium, but of sizes of approximately $0.5 \mu\text{m}$. Thus, it is reasonable that the interaction between Nafion molecules and Pt/C particles influences the formation of aggregates of Nafion molecules and Pt/C particles. For the purpose of evaluating the distribution of Pt/C and Nafion on the surface of the catalyst layer, the weight fractions of Pt and F were measured by EDS. Pt and F correspond to Pt/C particles and Nafion (perfluorsulfonic acid) molecules, respectively. The weight fraction can be used as a relative and quantitative value since it is not calibrated against a standard sample. The variations in the weight fractions of F and Pt are shown in Fig. 5. When a cluster or aggregate was observed on the surface, EDS measurements were conducted both on the flat unfractured surfaces and on the surface of fracture itself. The ratio of the weight fraction of F to that of Pt is plotted against the mixing time in Fig. 5 to allow comparison of the distributions of Nafion molecules and Pt/C particles for slurries of both compositions.

For the slurry with the lower solid volume fraction ($\phi = 0.018$), the fraction of Pt decreased and that of F increased gradually until they both reached steady values that were almost equal both on the flat surfaces and on the surface of aggregates. Thus, the ratio of the weight fraction of F to that of Pt largely remained constant after more than 4 h of mixing. The behaviors of the weight fractions of F and Pt at the initial stages of mixing were explained by the adsorption of Nafion molecules on the surfaces of the Pt/C particles, which were reported by Uchida et al. [17]. At the early stage of mixing, the catalyst particles, which did not connect with the Nafion molecules, form cluster. The catalyst particles and clusters settle down in a drying process and then Nafion will be concentrated on the surface of dried catalyst layer. As mixing proceeded, the catalyst particles were dispersed and stabilized by the adsorption of Nafion molecule. As a result, the weight fractions of F and Pt on the dried surface became stable and the composition will be same with that of the slurry. Therefore, the termination of Nafion adsorption can be judged both by the stable volume in small pore and by the stable composition on the surface. Additionally, it was found that the final ratio of the weight fractions of F to that of Pt on

the surface of aggregate (at 12 or 24 h of mixing) and on the flat surface showed almost the same value. It indicated that the aggregate was constructed by the Pt/C particles well connected with Nafion molecules after the mixing of a sufficient long duration.

However, in the catalyst layer produced from the slurry with insufficient Nafion compared with the amount of Pt/C particles ($\phi=0.035$), the fractions of F and Pt changed in an unexpected manner. At the initial stages of mixing, both fractions remained almost constant on flat areas. Much higher viscosity of the slurry (Fig. 2) implies the cluster of catalyst particles is difficult to move in slurry. Thus, the particle sedimentation in a coating was probably suppressed. As a result, the distribution of materials was homogeneous until the slurry became homogeneous and the viscosity increased again. After 4 h, the fraction of Pt decreased suddenly, whereas that of F increased gradually. At that time, the catalyst particles, which do not adsorb with Nafion, settled down and the decrease in the weight fraction of Pt on the surface was observed. The weight ratio of F to Pt on aggregates or clusters remained roughly constant independent of the mixing time and which was almost the same with that on flat surface at the initial stage of mixing. Uchida et al. [17] reported that Nafion intruded into the spaces between aggregates before entering the spaces within aggregates themselves during the mixing process. Thus, if the Nafion content is insufficient to cover each Pt/C particle, the ratio of the weight fractions on the flat surfaces and aggregates should finally be different. This is probably because both aggregates and clusters were formed with Nafion at the average composition of the slurry.

The results of elementary analysis suggest that the Nafion molecules were well dispersed and stabilized by adsorption on the Pt/C particles when the slurry contains sufficient Nafion. However, with insufficient Nafion, some of the Pt/C particles are not well attached with Nafion molecules even after attaining a homogeneous distribution. That is, after the dispersal of clusters, the uniformly distributed Nafion molecules are selectively attached to Pt/C particles and form aggregates. Therefore, for the slurry with inadequate Nafion, a long mixing time will increase the fraction of Pt/C particles that do not contribute to electricity generation. As seen in Fig. 2, the slurry with $\phi=0.035$ showed a stable viscosity from 2 to 8 h of mixing. We believe that this stable viscosity implies the adsorption of Nafion molecules on Pt/C particles and the resulting decrease in the fraction of free Nafion. Although the mechanism behind the subsequent increase in viscosity is not clear, we speculate that it may be due to the formation of aggregates covered with Nafion.

3.4. Electrochemical characteristics

The catalyst layers produced from the slurries mixed for different durations were used at the cathode, and the resulting cell performance was evaluated by plotting the I – V curve. The characteristic results for both slurry compositions are shown in Fig. 6. We found that for the slurry with $\phi=0.018$, the catalyst layers produced from the slurries mixed for 2 and 24 h showed almost the same performance. Furthermore, each characteristic value, i.e., apparent viscosity, small pore volume and F/Pt, became almost stable at that time. Thus, this slurry will achieve maximum performance after just 2 h of mixing. For the slurry with $\phi=0.035$, the cell voltage reduced with an increase in the mixing time. Since the decrease in voltage was clearly seen at high current densities due to proton transport, it is probably to the result of the insufficient amount of Nafion. EDS measurement showed that the distribution of the materials changed inhomogeneously after 4 h of mixing.

The variations in cell voltage at a current density of $50 \text{ A m}^{-2}\text{-Pt}$ are plotted against the mixing time in Fig. 7. For the slurry with

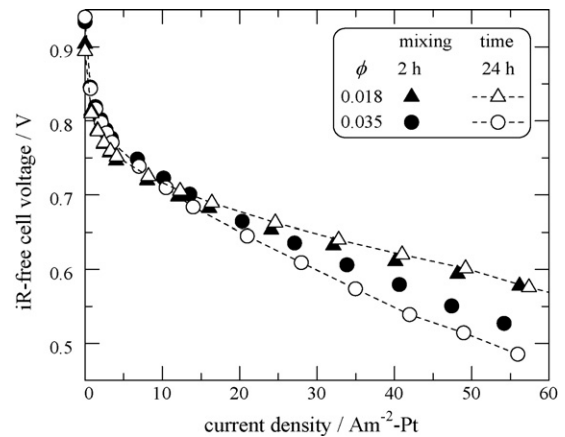


Fig. 6. I – V curves of cells with catalyst layers of different compositions.

$\phi=0.035$, at 0.5 h of mixing, a higher cell voltage was obtained than that from the slurry with $\phi=0.018$. At the early stage of mixing, however, the dispersal of Pt/C particles will be significantly scattered. Thus, the cell voltage at 0.5 h of mixing will have a possibility of reverse. As mixing progressed, the catalyst layers made from the concentrated slurry attained a steady state after more than 1 h of mixing. The mixing time required to obtain a stable performance is much shorter than that predicted from the physical or chemical characteristics described above. The difference may be caused by the change of the internal structure of the catalyst layer in a hot-press process. On the other hand, the slurry with $\phi=0.035$ also showed almost constant performance for more than 1 h of mixing, but we also observed a small drop in cell voltage after 4 h of mixing. The voltage drop must correspond to the change of Nafion distribution in a catalyst layer described in previous sections. Thus, we can obtain a clear agreement between the behaviors of apparent viscosity, small pore volume, surface material distribution, and cell voltage.

The pore structure and Nafion distribution must have changed during the hot-pressing process, as large pore could have been broken, allowing the entry of the Nafion from the ion-exchange membrane. Therefore, in future, elementary analysis and investigations of the pore size distribution and electrochemical characteristics must be performed both before and after the formation of the MEA.

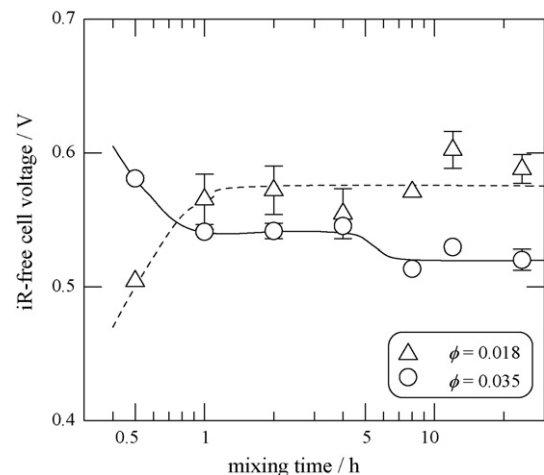


Fig. 7. Cell voltage at $50 \text{ A m}^{-2}\text{-Pt}$ as a function of mixing time.

4. Conclusion

The aim of this work is to clarify the changes in the structure of aggregate between Nafion molecules and Pt/C particles during the slurry preparation process and verify the relevance of the empirical criteria used to determine the completion of the mixing process. The viscosity of the slurry was measured to reveal details of structures of aggregates in the slurry. The structures of the catalyst layers were evaluated from the pore size distribution. Elementary analysis was conducted to determine the distribution of materials. Finally, a single cell test was used for electrochemical evaluation.

The results for the slurry with sufficient Nafion in relation to the Pt/C particle content were reasonable. That is, as mixing progressed, the Pt/C particles and Nafion molecules were dispersed homogeneously and Nafion molecules were adsorbed on the surface of the Pt/C particles. As a result, the viscosity, internal structure, elementary distribution, and cell performance changed equally with mixing time and could be stabilized by mixing for a sufficiently long period. On the other hand, for the slurry with insufficient Nafion, mixing for a sufficiently long duration will definitely afford a catalyst layer that produces stable but non-maximum cell performance. During the mixing process, Nafion molecules, which are first fully dispersed and subsequently adsorbed on the surfaces of aggregates of Pt/C particles, are consequently localized on these aggregates, drastically reducing the percolation of Nafion. Further, the fraction of Pt/C particles that do not contribute to electricity generation is increased.

Thus far, no clear evidence or information has been available to provide quantitative criteria for the slurry preparation process.

From the results of this work, we are convinced that the use of empirically determined criteria such as 'enough viscous' is fully reasonable. Furthermore, the viscosity of slurry can be a useful determiner of slurry preparation conditions in actual practice.

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