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Influence of Amount of Catalyst Formed on Carbon Support on Performance Characteristics of Fuel Cell with Carbon Nanotube-supported Catalyst

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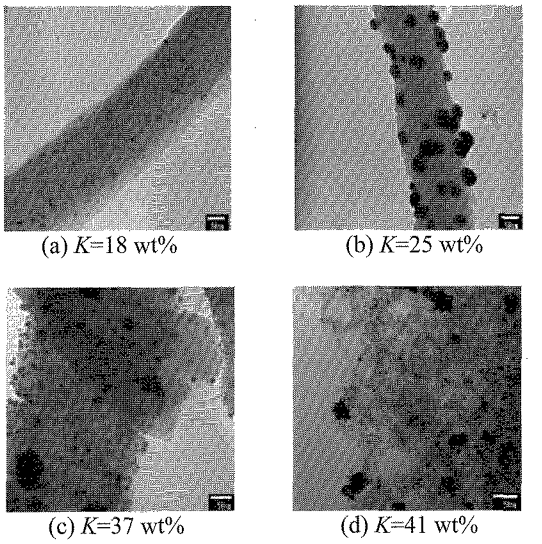
We experimentally investigated the influence of amount of catalyst formed on carbon nanotube support on the performance of polymer electrolyte fuel cell (PEFC). Platinum was used as the catalyst. We prepared four kinds of CNT's with Pt of 18 wt%, 25 wt%, 37 wt% and 41 wt%. First, we measured the optimum mass of electrolyte in the catalyst paste for each CNT-supported catalyst. Secondly, the output voltage and power density of the PEFC having the CNT-supported catalyst were estimated. It was found that the PEFC with CNT support of about 29wt%Pt could generate the highest electric power. Furthermore, the PEFC with CNT support may have lower optimum amount of Pt than that with carbon powder-supported catalyst which is widely used.
Key words: carbon nanotube, polymer electrolyte fuel cell (PEFC), catalyst, carbon support, performance characteristics

1. INTRODUCTION

Fuel cell is attracting much attention as an electric power generation method with low emission, high output density and efficiency[1]. Especially, the research and development task of polymer electrolyte fuel cell (PEFC) is now energetically pushed forward because it operates at relatively lower temperature and can be small as well as light. However, there are some needs for the performance for the practical use. An application of carbon nanotube (CNT) to the material supporting catalyst for the PEFC to improve the performance has been studied[2][3]. This time the influence of amount of catalyst formed on the CNT support on performance of PEFC was investigated experimentally. We prepared some CNT's having catalyst with different ratio.

2. CNT-SUPPORTED CATALYST

We used some CNT's having different amount of catalyst formed on them. We adopted Pt as the catalyst. In this paper, the ratio of the mass of Pt on the CNT to that of the CNT-supported catalyst was defined as K. We used the CNT-supported catalysts with K of 18 wt%, 25 wt%, 37 wt% and 41 wt%. Figure 1 shows the transmission electron microscope images of the CNT-supported catalysts. Figure (a), (b), (c) and (d) correspond to the images for the CNT-supported catalyst with K=18 wt%, 25 wt%, 37 wt% and 41 wt%, respectively. The black specks in Fig. 1 are Pt particles. There is difference in the condition of Pt on the CNT. In case of K=18 wt%, Pt dispersed well on the CNT. However, it cannot be said that the dispersibility of the other catalysts was good. Table 1 summarizes the specifications of each CNT-supported catalyst. There were some differences in the specifications.



(a) K=18 wt% (b) K=25 wt% (c) K=37 wt% (d) K=41 wt%

Fig. 1. Transmission electron microscope photograph of CNT-supported catalyst.

Table 1. Specifications of CNT-Supported Catalyst.

K	18 wt%	25 wt%	37 wt%	41 wt%
Pt size	2-20nm	10-50nm	2-50nm	10-100nm
CNT diameter	30-200nm	30-200nm	40-400nm	10-50nm
CNT length	1-10μm	1-10μm	0.5-5μm	1-50μm

3. MIXTURE RATIO IN CATALYST PASTE

The catalyst paste was made by mixing the CNT-supported catalyst and electrolyte solution and applied on diffusion electrodes of the PEFC. Nafion^{TM} was used as the electrolyte. The mixture ratio of Nafion^{TM} solution to CNT-supported catalyst can affect the performance of the PEFC. Hence, it is important to grasp the optimum mixture ratio in order to obtain the large electric power output. We experimentally investigated the optimum Nafion^{TM} mass per unit section of electrode. The amount of Pt on the electrode was 1.0 mg/cm^2. We made membrane electrode assemblies (MEA's) using the CNT-supported catalysts of 18 wt%, 25wt% and 41wt% and tested their performance. The cross section of the electrode was 2.2 mm × 2.2 mm (4.84 mm^2) and Nafion^{TM} 115 (thickness: 127 μm) was used as the electrolyte membrane.

Figure 2 shows the measured voltage-current density characteristics in case of K=25 wt%. The results for MEA's with different Nafion^{TM} mass (0.5, 1.1, 1.5 and 2.1 mg/cm^2) are indicated together in this figure. In the experiments, the volumetric flow rates of H_2 and O_2 were 300 ml/min and 150 ml/min. The gas pressure was atmospheric pressure and the cell temperature was 70 ℃. As seen in Fig. 2, the voltage for 1.5 mg/cm^2 is highest among the four conditions. Figure 3 describes measured results the power density as a function of current density. The power density-current density characteristics have a maximum value. The maximum power density under the condition of Nafion^{TM} mass of 1.5 mg/cm^2 was largest and the value was 0.46 W/cm^2.

Figure 4 indicates the maximum power density as a function of Nafion^{TM} mass. The characteristics also have the maximum value. This is because the effective three phase contact cannot be formed in case that the electrolyte is too much as well as too little[1]. In this paper, the condition under which the maximum value of the power density was highest was the optimum one. Hence, Nafion^{TM} mass of 1.5 mg/cm^2 was the optimum condition in case of K=25 wt%.

We measured the optimum Nafion^{TM} mass for the MEA's with the other K's by the same method. The results are shown by the mark "□" in Fig. 5. The optimum mass decreased with an increase in K. In case of smaller K, we need more CNT with Pt, i.e., the catalyst layer is thicker because the amount of Pt on the electrode was specified to 1.0 mg/cm^2. Hence, more Nafion^{TM} is needed.

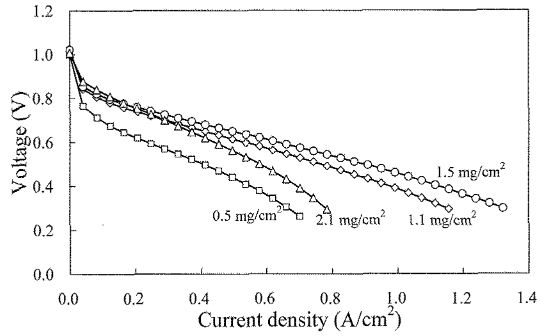


Fig. 2. Voltage-current density characteristics of PEFC with CNT-supported catalyst (K=25 wt%, H_2: 300 ml/min, O_2: 150 ml/min, gas pressure: atmospheric, cell temperature: 70 ℃).

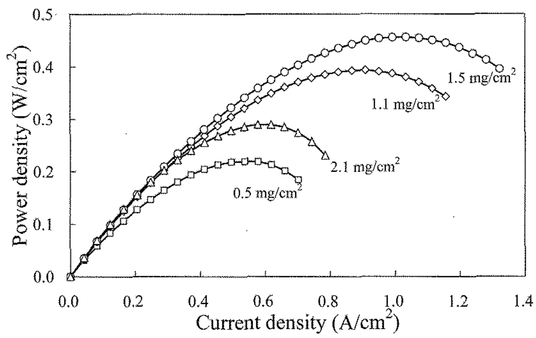


Fig. 3. Power density-current density characteristics of PEFC with CNT-supported catalyst (K=25 wt%, H_2: 300 ml/min, O_2: 150 ml/min, gas pressure: atmospheric, cell temperature: 70 ℃).

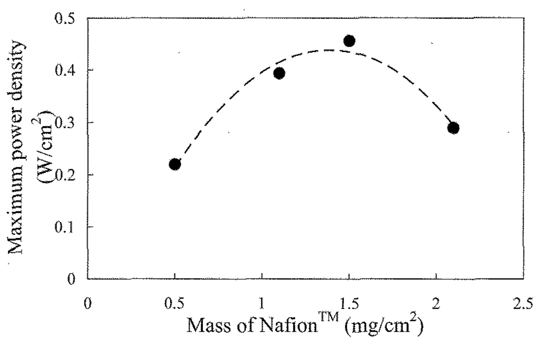


Fig. 4. Maximum power density as a function of Nafion^{TM} mass in catalyst paste (K=25 wt%).

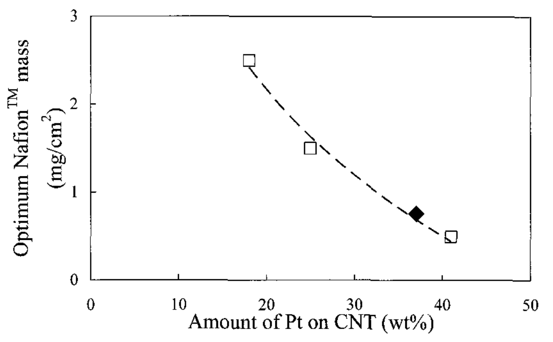


Fig. 5. Optimum Nafion™ mass as a function of ratio K of Pt formed on CNT support.

4. PERFOEMANCE OF PEFC WITH CNT-SUPORTED CATALYST

4.1 Performance Characteristics

Figure 6 and 7 show the voltage-current density characteristics and the power density-current density characteristics of MEA's with the CNT-supported catalyst having different K. We made the MEA's considering the optimum Nafion™ mass mentioned in chapter 3. The Nafion™ mass for the CNT-supported catalyst of K=37 wt% was decided by interpolation in Fig. 5. The mass of Nafion™ in case of K=37 wt% is shown by the mark "◆" in Fig. 5. The measurement conditions were the same as those in experiments mentioned in previous chapter. Form Fig. 6, the voltage for K=25 wt% is highest of the four conditions. As seen in Fig. 7, maximum power density for K=25 wt% is also largest. The Value of the maximum power density was 0.46 W/cm^2. Figure 8 describes the maximum power density as a function of K by the mark "●". As seen in this figure, the characteristics have the peak value. In case of small K, the catalyst layer can be thicker and the electric conductivity and gas permeability can be worse. On the other hand, the effective surface of Pt may decrease in case of too large K. Hence, there is a certain K under which the maximum power density takes the peak value. The quadratic approximation curve is also shown by solid line in Fig. 8. From this expression, K in case that the highest power density is generated can be decided to be 29 wt%.

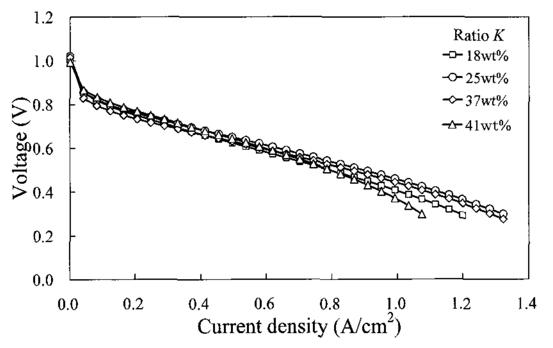


Fig. 6. Voltage-current density characteristics of PEFC with CNT-supported catalyst (H_2: 300 ml/min, O_2: 150 ml/min, gas pressure: atmospheric, cell temperature: 70 ℃).

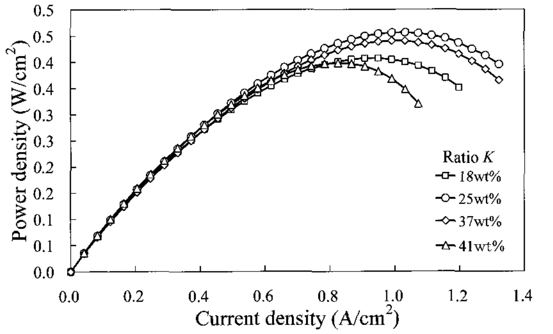


Fig. 7. Power density-current density characteristics of PEFC with CNT-supported catalyst (H_2: 300 ml/min, O_2: 150 ml/min, gas pressure: atmospheric, cell temperature: 70 ℃).

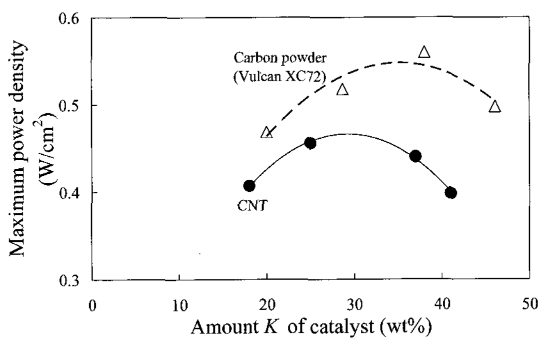


Fig. 8. Maximum power density as a function of amount K of catalyst formed on carbon nanotube.

4.2 Comparison with Carbon Powder

Carbon powder is in wide use as a support material for the catalyst in PEFC. The performance characteristics of the MEA with carbon powder were estimated for comparison. In this study, Vulcan™ XC72 was used as the support carbon. The results for carbon powder were also indicated in Fig.8. As seen in this figure, K for carbon powder when the power density was maximal was 35 wt%. This value was larger than that for the CNT. In case of carbon powder, the generated current flows through innumerable contact points between the carbon particles although the current can conduct along the CNT in case of CNT-supported catalyst. This means that the electric resistance in catalyst layer can be larger in the carbon powder case than the CNT case. Furthermore, the space between carbon particles may be narrower than that between the CNT's because the particle size of the carbon powder is smaller than that of the CNT. So, the gas permeability may be better in case of the CNT. On the other hand, the effective surface of the CNT is smaller than that of carbon powder. That is, it is harder to disperse Pt on the CNT than the carbon powder. As a result, K for carbon powder when the power density gets maximized is larger than that for CNT. This may be the remarkable property of the CNT-supported catalyst.

5. CONCLUSIONS

Influence of the amount of Pt catalyst on CNT support on the electric power generation characteristics was experimentally investigated. First, the optimum value of Nafion^{TM} mass in catalyst paste. Secondly, the performance of the PEFC with CNT-supported catalyst was estimated. When the CNT-supported catalyst of about 29 wt% was used, the power density took the peak value. This value of catalyst amount was smaller than that for carbon powder-supported catalyst.

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