



Short communication

# Stainless steel bipolar plate coated with carbon nanotube (CNT)/polytetrafluoroethylene (PTFE) composite film for proton exchange membrane fuel cell (PEMFC)

Yoshiyuki Show\*, Kenta Takahashi

Department of Electrical and Electronic Engineering, Tokai University, 1117 Kitakaname, Hiratsuka, Kanagawa 259-1292, Japan

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## ABSTRACT

Composite film of carbon nanotube (CNT) and polytetrafluoroethylene (PTFE) was successfully formed by using their dispersion fluids. This CNT/PTFE composite film was electrically conductive in the range of  $10\text{ S cm}^{-1}$ . The proton exchange membrane fuel cell (PEMFC) was assembled with the stainless steel bipolar plate coated with the CNT/PTFE composite film. This coating decreased the contact resistance between the surface of the bipolar plate and the membrane electrode assembly (MEA). Therefore, the output power of the fuel cell increased by 1.6 times.

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## 1. Introduction

The proton exchange membrane fuel cell (PEMFC) is an ideal candidate for automotive propulsion application, because it has high efficiency and cleanliness as a power source. Graphite bipolar plate has excellent natures in terms of chemical stability and electrical conductivity. However, it contributes significantly to the weight and cost of PEMFC although automotive application requires miniaturization and low cost. Metals such as stainless steel have been considered as alternative materials to graphite bipolar plate, because they are produced using chemical etching or press processes. Moreover, they are generally thinner than graphite bipolar plates thus resulting in a fuel cell stack that is both light in weight and small in volume.

An ideal bipolar plate material should be corrosion-resistant, because pH in fuel cell has a low value of 3.7–4.1 [1]. However, metal bipolar plates were corroded in the strong acidic environment in PEMFC. Therefore, carbon-based [2–5] and metal-based [6] coating on metal bipolar plates has been studied to improve corrosion resistance and conductivity. Although amorphous carbon film coating on the metal bipolar plates increases the output power of fuel cell [2,3], this technique is far from the practical application,

because its coating cost using the chemical vapor deposition (CVD) method is of high value.

In this study, electrically conductive composite film consisting of carbon nanotube (CNT) and polytetrafluoroethylene (PTFE) was coated on stainless steel bipolar plates. Moreover, the contact resistance between the surface of the bipolar plate and the membrane electrode assembly (MEA), and the output power of the fuel cell were investigated.

## 2. Experiments

### 2.1. Formation of CNT/PTFE composite film

CNT/PTFE composite film was formed from CNT and PTFE dispersion fluids. The CNT dispersion fluid was made from multi-walled type CNTs. Cellulose derivatives were added to the water to disperse the CNT. This CNT dispersion was made by means of the technique reported in Ref. [7]. The CNT concentration in the dispersion fluids was 3%. The CNTs were well-dispersed in the water even when the CNT concentration was of a high value. Water-based commercial PTFE dispersion was used in this study. The PTFE particles in the water were 0.20–0.40  $\mu\text{m}$  in diameter. The PTFE concentration was 60%. The separate CNT and PTFE dispersion fluids were mixed and stirred by ultrasonic wave application. The CNT concentration in this mixed fluid was varied from 0 (pure-PTFE dispersion) to 100% (pure-CNT dispersion). An additional separate

\* Corresponding author. Tel.: +81 463 58 1211; fax: +81 463 59 4014.

E-mail address: [show@keyaki.cc.u-tokai.ac.jp](mailto:show@keyaki.cc.u-tokai.ac.jp) (Y. Show).

dispersion fluid, formed from PTFE dispersion fluid and CNT powder instead of CNT dispersion fluid, was also prepared as a reference. The CNT/PTFE dispersion was applied at a 50  $\mu\text{m}$  thickness to a glass substrate or stainless steel bipolar plate. The sample was dried in a 40  $^{\circ}\text{C}$  atmosphere for 30 min and then was heated at 350  $^{\circ}\text{C}$  for 10 min.

Polarization tests were carried out in 1 M  $\text{H}_2\text{SO}_4$  from corrosion potential to 1800 mV vs. Ag/AgCl for stainless steel plate (304SS) coated with the CNT/PTFE composite film. The Ag/AgCl electrode was used as the reference. The scan rate of the potential voltage was 5 mV  $\text{s}^{-1}$ .

## 2.2. Fabrication of fuel cell using metal bipolar plates coated with the CNT/PTFE composite film

Polymer electrolyte membrane fuel cells (PEMFC) were assembled by using bare stainless steel (304SS) and the CNT/PTFE composite film coated stainless steel bipolar plates. The concentration of the CNT in the coated film is 75%. Nafion 117 was used as the electrolyte. Platinum particles with a density of 0.25  $\text{mg cm}^{-2}$ , which were supported on acetylene black powder, were used as the catalyst for the anode and cathode electrodes. The size of the MEA (membrane electrode assembly) was 4 cm  $\times$  4 cm. Humidified hydrogen and oxygen gases flowed into the anode and the cathode at 100 and 200 sccm respectively. The fuel cell was operated at R.T.

## 3. Result and discussion

Fig. 1 shows the mixed dispersion fluids of CNT and PTFE made by two kinds of the methods. Fig. 1(a) shows the fluid which made by mixing the powdered CNT and the PTFE dispersion as a reference. When the powdered CNT was used, both the CNT and the PTFE were settled to the bottom of the bottle. Clear supernatant fluid was appeared at the top. Fig. 1(b) shows the dispersion fluid made by mixing dispersion fluids of the CNT and the PTFE. The CNT was well-dispersed in the PTFE dispersion. The CNT was not settled to the bottom of a bottle for over 1 week. This dispersion was used to form the CNT/PTFE composite film in the following experiments.

Fig. 2 shows the TEM image for the CNT/PTFE composite film at the CNT concentration of 25%. The CNTs were uniformly distributed throughout the film. The CNT in the film was approximately 10 nm in diameter. The TEM image showed that CNTs touch each other within the film.

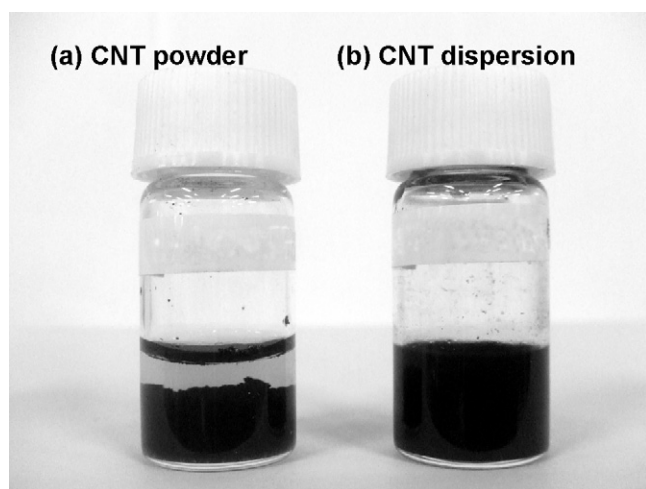


Fig. 1. (a) The fluid made by mixing the powdered CNT into the PTFE dispersion as a reference. (a) The fluid made from dispersions of the CNT and the PTFE. The CNT was well-dispersed in the PTFE dispersion.

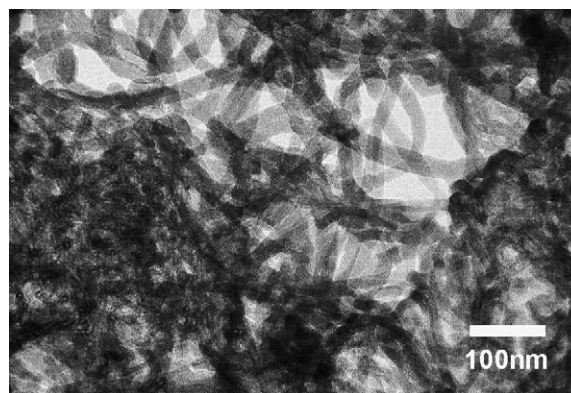


Fig. 2. TEM image of the CNT/PTFE composite film.

Fig. 3 shows the electrical conductivity of the CNT/PTFE composite film at various CNT concentrations. No electrical conduction was observed for the pure PTFE film, since PTFE is a highly insulating material. The composite film at the CNT concentration of 5% showed an electrical conduction of 0.1  $\text{S cm}^{-1}$ . The electrical conductivity was drastically increased by 10 times with an increase in the CNT concentration of the film to 10%. The electrical conduction linearly increased to the order of 20  $\text{S cm}^{-1}$  with an increase in the CNT concentration from 10 to 100%. The films with a CNT concentration below 90% have flexibility, because the CNTs are bound by the PTFE. On the other hand, the film formed with exclusively the CNT is not flexible. This film was easily broken into small particles of CNT by scratching it with tweezers. A water-resistance test was also carried out. The composite films below the CNT concentration of 90% did not break-up in water. However, the film where no PTFE had been added (100% CNT) was redispersed in water.

The CNTs were successfully dispersed in the composite film by using the CNT dispersion fluid. The CNTs work as a conductive path throughout the composite film. A three-dimensional conductive network of CNTs is needed to form a highly conductive film. A CNT concentration of 5% is not high enough to form the three-dimensional conductive networks. Therefore, a 5% concentration film has a relatively low conductivity value of 0.1  $\text{S cm}^{-1}$ . The conductivity of the film drastically increases from 0.1 to 2  $\text{S cm}^{-1}$  with an increase in the CNT concentration up to 10%, this is because

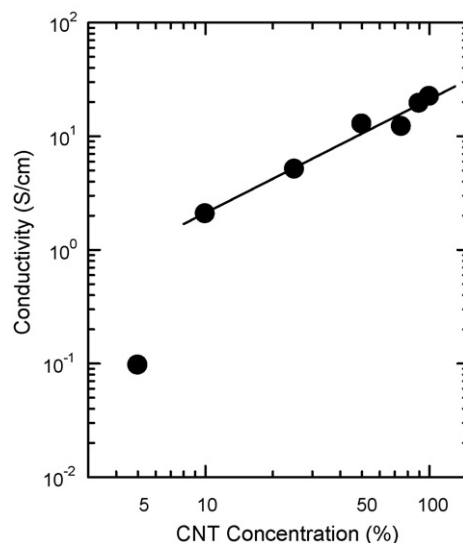


Fig. 3. The electrical conductivity of the CNT/PTFE composite film at various CNT concentrations.

**Table 1**  
Contact resistance between surface of bipolar plate and carbon paper.

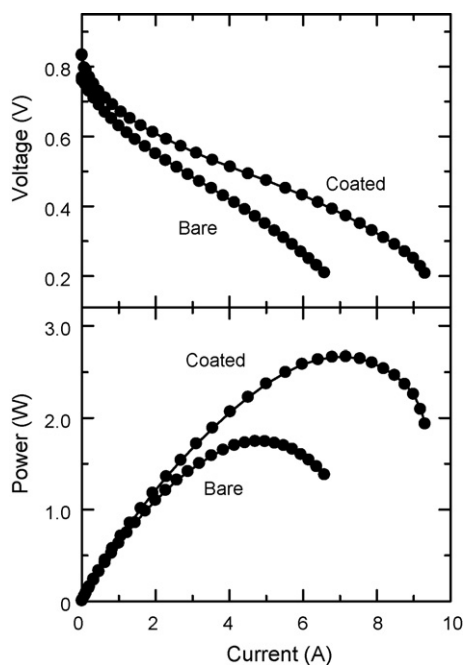
Bipolar plate	Contact resistance ( $\text{m}\Omega \text{ cm}^{-2}$ )
Bare	46
Coated	12

the three-dimensional conductive network is formed. This drastic increase in conductivity is called the percolation threshold [8]. The conductivity of the film linearly increases with the CNT concentration above the percolation threshold of 10%.

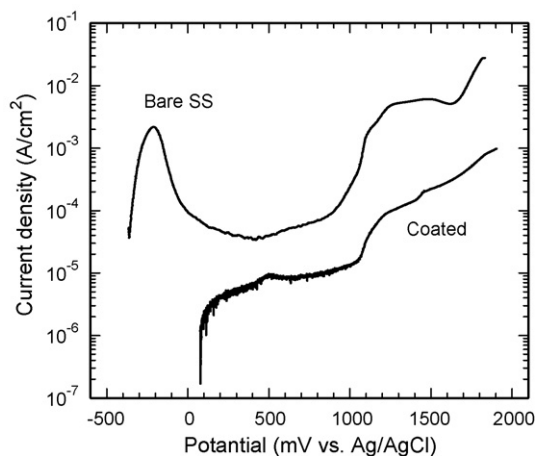
The composite film at the CNT concentration of 75% was applied to bipolar plates of the proton exchange membrane fuel cell. The contact resistance between the surface of bipolar plate and carbon paper, which has been used as gas diffusion layer in the MEA, was measured. In this study, the carbon paper was sandwiched between two pieces of the bipolar plate. The contact resistance was measured by flowing current between bipolar plates through the carbon paper. Table 1 shows the results of the contact resistance measurement. When the bare stainless steel bipolar plates were used, the contact resistance to the carbon paper was  $46 \text{ m}\Omega \text{ cm}^{-2}$ . On the other hand, the stainless steel bipolar plate coated with the composite film showed lower contact resistance of  $12 \text{ m}\Omega \text{ cm}^{-2}$ .

Fig. 4 shows the dependence of the current on the output voltage and the electric power for the fuel cells assembled with the bare stainless steel bipolar plates and the bipolar plates coated with the CNT/PTFE composite film. Both of fuel cells showed an open voltage of 0.8 V. No dependence on surface coating to the bipolar plate on the open voltage was observed. The output voltage decreased with an increase in the current, because of the IR drop. The IR drop of the fuel cell, using the bipolar plates coated with the composite film, is lower than that of the bare bipolar plates.

The maximum output power for the fuel cell using the bare bipolar plates was 1.7 W. On the other hand, the fuel cell using the bipolar plates coated with the composite film showed a maximum output power of 2.7 W, which is higher than that of the bare bipolar plates by 1.6 times.



**Fig. 4.** The dependence of the current on the voltage and the electrical power generated by the polymer electrolyte membrane fuel cells (PEMFC). The fuel cells were assembled using either bare stainless steel bipolar plates or bipolar plates coated with the CNT/PTFE composite film.



**Fig. 5.** Polarization curves for bare stainless steel plate and stainless steel plate coated with the CNT/PTFE composite film.

Fig. 5 shows polarization curves for bare stainless steel plate and stainless steel plate coated with the CNT/PTFE composite film (CNT concentration of 75%). The bare stainless steel showed the corrosion potential of 365 mV vs. Ag/AgCl. The strong peak of active–passive transition appeared at 200 mV vs. Ag/AgCl. The passive current with  $5 \times 10^{-5} \text{ A cm}^{-2}$  was observed at 600 mV vs. Ag/AgCl. On the other hand, the stainless steel plate coated with the CNT/PTFE composite film showed the passive current with  $8 \times 10^{-6} \text{ A cm}^{-2}$  which was lower than that of the bare stainless steel. This polarization curve test implies that coating of the CNT/PTFE composite film on stainless steel improves corrosion resistance.

The coating of the CNT/PTFE composite film decreases the contact resistance between the bipolar plate and the carbon paper, because this film has high electrical conductivity. The decrease in the contact resistance leads to the inner resistance of the fuel cell to be decreased. Therefore, the output power of the fuel cell was increased by this coating.

The surface of stainless steel bipolar plates was corroded during generation of electricity, because the inside of the fuel cell has strong acidic environment [1]. The surface corrosion increases the contact resistance between the bipolar plate and the MEA and deteriorates the performances of MEA due to contamination of Pt catalyst with dissolved metal ions. Therefore, the output power of the fuel cell is decreased. The polarization test in this study suggests that the CNT/PTFE composite film protects the surface of the bipolar plates against the corrosion. Therefore, the coating of the composite film on bipolar plates seems to increase the lifetime of the fuel cell.

#### 4. Summary

The CNT/PTFE composite film was formed by using the dispersion fluids of the CNT and the PTFE. This film showed properties of electrical conduction facilitated by the CNTs within the film. The conductivity of the composite film increased with an increase in the CNT concentration. The film having the conductivity of  $12 \text{ S cm}^{-1}$  was observed at the CNT concentration of 75%. The fuel cell assembled with the stainless steel bipolar plates, which was coated with this composite film, showed higher output power than that of bare bipolar plates by 1.6 times.

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## References

- [1] A. Pozio, R.F. Silva, M. De Francesco, L. Giorgi, *Electrochim. Acta* 48 (2003) 1543.
- [2] Y. Show, *Surf. Coat. Technol.* 202 (2007) 1252–1255.
- [3] Y. Show, M. Miki, T. Nakamura, *Diamond Relat. Mater.* 16 (2007) 1159–1161.
- [4] T. Fukutsuka, T. Yamaguchi, S.I. Miyano, Y. Matsuo, Y. Sugie, Z. Ogumi, *J. Power Sources* 174 (2007) 199–205.
- [5] C.Y. Chung, S.K. Chen, P.J. Chiu, M.H. Chang, T.T. Hung, T.H. Ko, *J. Power Sources* 176 (2008) 276–281.
- [6] V. Mehta, J.S. Cooper, *J. Power Sources* 114 (2003) 32–53.
- [7] N. Minami, Y. Kim, K. Miyashita, S. Kazaoui, B. Nalini, *Appl. Phys. Lett.* 88 (2006) 93123.
- [8] S. Kirkpatrick, *Rev. Mod. Phys.* 45 (1973) 574.