Electroless Ni-plating on PTFE fine particles

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Abstract

A Ni electroless plating process was used with polytetrafluoroethylene (PTFE) fine particles (25–500 μm). Using nonionic hydrocarbon surfactant, PTFE particles were dispersed in the plating bath. The PTFE hydrophobicity was sufficiently high that Ni was deposited partly on the PTFE surface in the initial step. The Ni-PTFE particles were formed into the Ni-PTFE plate by heat treatment at 350 °C after pressing. The Ni-PTFE plate had electrical conductivity and gas permeability, which were influenced by the pore distribution in the plate. Pores with 1 μm diameter might be especially important to impart high gas permeability to the Ni-PTFE plate.

1. Introduction

Recently, polymer electrolyte fuel cells (PEFCs) have been studied for use as power sources for electric vehicles, on-site electric generators, and mobile electronics devices. For them, carbon materials have generally been used as separators. Carbon separators have superior corrosion resistance to that of metal separators, but they present an important problem: their electrical conductivities are lower than those of metal separators. Especially, producing a channel on the separator for reactive gas passage entails high processing costs. Development of metal or carbon-composite (the carbon powder-resin composite) separators is being attempted to achieve low-cost processing [1].

Our research into formation of composite metal films on various polymer particles using electroless plating has progressed. Such films impart many features such as electrical conductivity, water-repellency, and plasticity. Metal-plated polymer particles must be freely shaped by pressing. Press-processing of the metal-plated polymer particles is necessary to

form optimal shapes for electrical conductivity and gas permeability. For this study, we attempted to produce an electrode for PEFC with reaction gas paths and a diffusion layer. Because polytetrafluoroethylene (PTFE) has high chemical stability and plasticity, we selected PTFE as the core material of polymer particles used in this study.

The PTFE particles must be hydrophilized to disperse them in an electroless plating solution because PTFE shows great hydrophobicity [2–5]. For hydrophilic treatment to impart hydrophilicity on the PTFE particles' surface, we used surfactants as the favored method. Surface modification of PTFE fine powder using a surfactant is more convenient than other methods such as electron beam irradiation and corona discharge. Generally, when the PTFE was rendered hydrophilic using the surfactant, a surfactant containing fluorine (fluorochemical surfactant) was used because the perfluoro group in it exhibits high adsorbability against PTFE. However, fluorochemical surfactants are not a popular commercial product and are classified as hazardous substances. In addition, some pyrolysis products must necessarily remain when sintering at 350 °C to produce an electrode. On the other hand, many kinds of hydrocarbon surfactants have been produced commercially. Hydrocarbon surfactants present environmental safety and cost advantages.

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This paper reports the hydrophilic treatment of PTFE particles using various surfactants, activating treatment for electroless plating, Ni-plating processes, and press-heating of Ni-plated particles to form an electrode plate for PEFC.

2. Results and discussion

2.1. Hydrophilizing PTFE particles using various hydrocarbon surfactants

To examine the creation of hydrophilicity on PTFE by treatment with the surfactant, various nonionic hydrocarbon surfactants were tested considering their hydrophilic lipophilic balance (HLB, Eq. (1)) [6], which is generally used as an index of a surfactant's character.

$$HLB = \frac{\text{Weight \% of EO group in molecule}}{5} \qquad (1)$$

Polyoxyethylene alkyl ethers having different numbers of ethylene oxide (EO) groups and different normal chain alcohol were used as surfactants in this study. The PTFE particles were dispersed in water with 0.2 wt% of 11 kinds of hydrocarbon surfactants. Among them, six reagents, $C_{12}H_{25}–O–(C_2H_4O)_2–H$ (BL-2), $C_{12}H_{25}–O–(C_2H_4O)_{4.2}–H$ (BL-4.2), $C_{16}H_{33}–O–(C_2H_4O)_2–H$ (BC-2), $C_{18}H_{37}–O–(C_2H_4O)_2–H$ (BS-2), $C_{18}H_{35}–O–(C_2H_4O)_2–H$ (BO-2), and $C_{18}H_{35}–O–(C_2H_4O)_7–H$ (BO-7V) were particularly effective to hydrophilize PTFE particles. The PTFE particles were dispersed easily into ionic aqueous solutions such as an electroless plating bath, a sensitizing bath, and activating bath using those surfactants. Fig. 1 portrays the contact angle of the water droplet of the nonionic-surfactant-treated PTFE surface and the HLB of the surfactant treated using various surfactants. The contact angles of the PTFE plates treated using the six surfactants described above were smaller, 5° or more, than the untreated PTFE (Blank) shown by t7he dashed line. On the other hand, the contact angles of PTFE treated using other nonionic surfactants, anionic surfactant, and cationic surfactants were almost equal to the contact angle of Blank. The nonionic surfactant of HLB < 10

imparted hydrophilicity to the PTFE [7]. The clouding point of each surfactant of HLB < 10 – the temperature at which the surfactant solution becomes milky from its original transparent state when the solution temperature is raised – was below room temperature because these surfactant's respective hydrophobicities were greater than their hydrophilicities. Because of the surfactant's hydrophobicity, the surfactant adsorbed to the PTFE surface through interaction with the highly hydrophobic PTFE surface. Thereby, it imparted hydrophilicity to the PTFE surface. On the other hand, the anionic, cationic, and high-HLB nonionic surfactants did not adsorb onto the PTFE surface because of their high hydrophilicity: they imparted no hydrophilicity to the PTFE surface. Fig. 1 shows that the surfactants can be categorized into two groups: a high hydrophilicity group with low contact angle, and a low hydrophilicity group with a slightly lower contact angle than that of Blank. The surfactant-treated PTFE with lauryl or oleyl group exhibited a low contact angle; hydrophilicity is affected not only by HLB. For example, comparing the surfactants with stearyl (BS-2) and oleyl (BO-2) groups having similar HLB values, BO-2 was superior in imparting hydrophilicity to PTFE. The hydrophilicity of the oleyl group is far superior to that of the stearyl group [8]. Hydrophilicity is apparently influenced not only by the HLB value, but also by the nature of the alkyl groups in surfactants.

Fig. 2 portrays the distribution of the number of EO groups analyzed using high-performance liquid chromatography (HPLC) in the commercially supplied nonionic surfactant $C_{12}H_{25}–O–(C_2H_4O)_2–H$ (BL-2). Distribution of additional ethylene oxide molecules occurs because the nonionic surfactant is usually synthesized by adding EO to alcohol. In this case, results showed that BL-2 is the mixture of the components with C_{EO} values from 0 to about 10. The contents of the components with C_{EO} of more than 11 have the value less than 0.5% which is negligible. HLB was evaluated by using the average of C_{EO} values (2 in this case). Fig. 1 shows that the number of EO groups in the nonionic surfactant with the same alkyl group greatly influenced the contact angle of the water droplet on the nonionic-surfactant-treated PTFE surface. To elucidate the influence of the additional number of EO
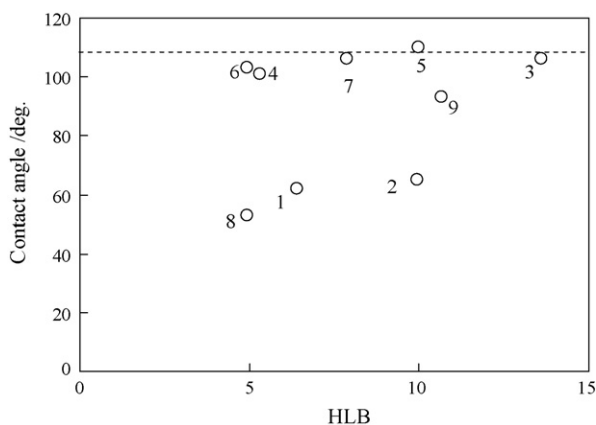

Fig. 1. Relationship between the contact angle of a water droplet on surfactant-treated PTFE and HLB of the surfactant: 1, BL-2; 2, BL-4.2; 3, BL-9EX; 4, BC-2; 5, BC-5.5; 6, BS-2; 7, BS-4; 8, BO-2V; 9, BO-7V. The dashed line at 109° shows to the blank (without surfactant) sample.
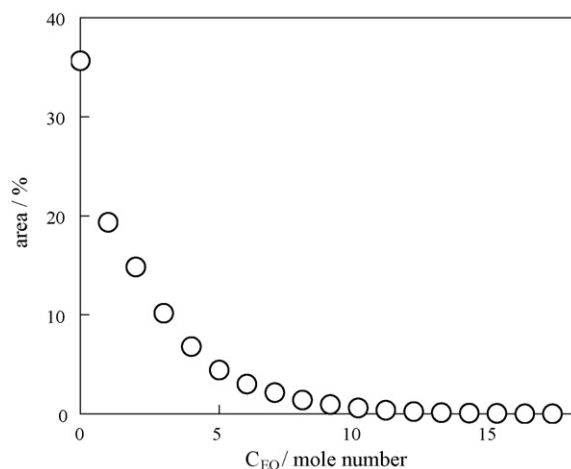

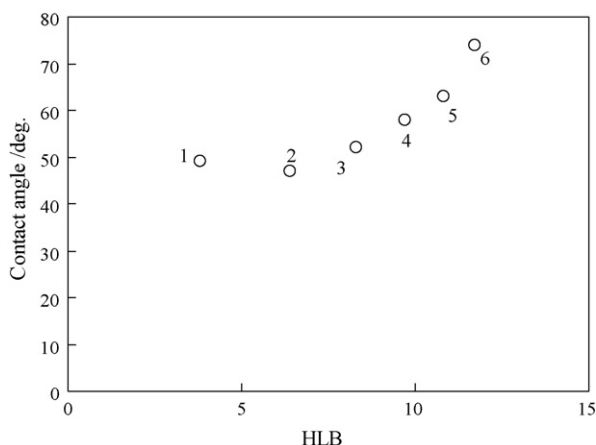Fig. 2. Added EO mol distribution of BL-2.

Fig. 3. Relationship between the contact angle of a water drop on nonionic-surfactant-treated PTFE and HLB of the surfactant: 1, BL-1SY; 2, BL-2SY; 3, BL-3SY; 4, BL-4SY; 5, BL-5SY; 6, BL-6SY.
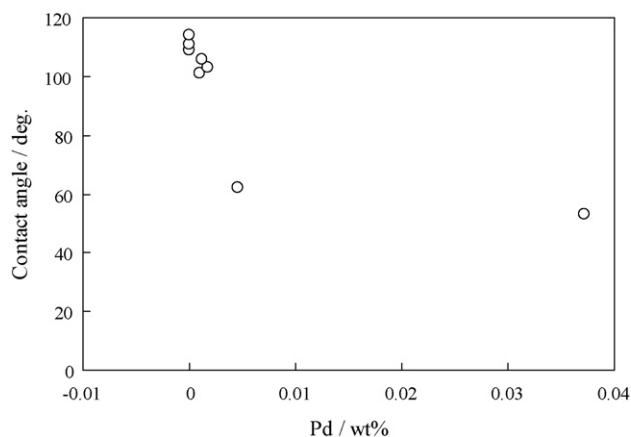

Fig. 4. Relationship between the contact angle a water drop on surfactant-treated PTFE and of Pd content: 1, blank; 2, CA-2150; 3, SLS; 4, BL-2; 5, BC-2; 6, BS-2; 7, BS-4; 8, BO-2.

molecules in the nonionic surfactant with the same alkyl group, the water droplet contact angle on a nonionic-surfactant-treated PTFE surface without influence from the distribution of EO number was measured. We experimented using a homogeneous polyoxyethylene nonionic surfactant, which had no distribution of the EO number. Fig. 3 shows that result. The contact angle increased with increasing HLB and the additional number of EO molecules more than two. The amount of the surfactant adsorbed on the PTFE surface may decrease with increasing the additional number of EO molecules. It may cause the contact angle approaches to the one of the original PTFE. On the other hand, the contact angle for the additional number of EO molecules of 1 was slightly larger than that of 2. This seems to be caused by the higher hydrophobicity of the surfactant with the additional number of EO molecules of 1. Therefore, we infer that the hydrophilic ability to the nonionic-surfactant-treated PTFE depends directly on the additional number of EO molecules in the nonionic surfactant with the same alkyl group. Moreover, because the commercially supplied nonionic surfactants have the additional distribution of EO molecules, the hydrophilicity related to the nonionic-surfactant-treated PTFE was affected by the distribution state of additional EO molecules in the nonionic surfactant.

2.2. Effect of hydrophilic treatment with various nonionic hydrocarbon surfactants on sensitizing with Sn(II) and activation with Pd(II)

Both Sn-sensitizing and Pd-activation activated the surfactant-treated PTFE particles for electroless Ni-plating. Fig. 4 shows the relationship between the Pd amount and the contact angle of a water droplet on the surfactant-treated PTFE particle. The contact angle was related to the Pd concentration. The PTFE particle that was hydrophilized in conditions under which the PTFE plate exhibited a low contact angle had large Pd content.

To discuss the role of surfactant in the activation process, it is important to consider the location at which Pd was deposited. The surfactant does not adsorb on PTFE surface homo-

geneously by its hydrophobic group like a micelle; instead, it forms a domain in which the surfactant is condensed without orientation because the HLB value of the surfactant here is so small that it is very difficult for it to have compatibility with water. This domain might be scattered on the PTFE surface. Apparently, PTFE can be wetted with water when the cover ratio of surfactant on PTFE reaches a certain value. Generally, the terminal group of PTFE was not often blocked by –CF$_3$; almost all were of the high polarity terminal group: –COF, –COOH, –CF=CF$_2$, –CONH$_2$, –CH$_2$OH, –CF$_2$H, –CF$_2$C$_2$H$_5$, etc. [9] Therefore, it seemed that the hydrophilicity of the terminal group of PTFE was higher than that of other PTFE hydrophobic surfaces, which were –CF$_2$–. This heterogeneous PTFE surface might reflect the non-uniform adsorption of surfactants.

During the Sn-sensitizing and Pd-activation processes, Sn(II), which is a reductant of Pd(II) and is hydrated in an aqueous solution, had to approach the hydrophilic domain on PTFE particle surface. Actually, Sn(II) in the hydrophilic domain might be oriented to F by the electrostatic interaction with Fδ- on the PTFE surface.

In addition, Pd ion must approach the hydrophilic domain on the PTFE surface and then be deposited as a fine colloidal particle of Pd metal. The Pd is expected to deposit on the PTFE surface directly or through the surfactant. Because the surfactant on the PTFE surface is quite soluble in ethanol, we sought to remove the surfactant on the PTFE surface after the activation process by washing it with ethanol. The Pd on the PTFE surface remained after washing with ethanol. Therefore, we infer that Pd was deposited on the PTFE surface directly, not through the surfactant.

Fig. 5(a) depicts a scanning electron microscopy (SEM) micrograph of the Ni-plated surface of PTFE particle with 0.53 wt% Ni contents. The Pd deposited as a catalyst was so fine that it was only slightly observed directly. However, the surface morphology of Ni-plated PTFE was influenced by the situation of Pd deposition: Ni was deposited partly on the PTFE surface, as shown in Fig. 5(a) in the initial stage of the deposition; it resembles a grain boundary on a polycrystalline
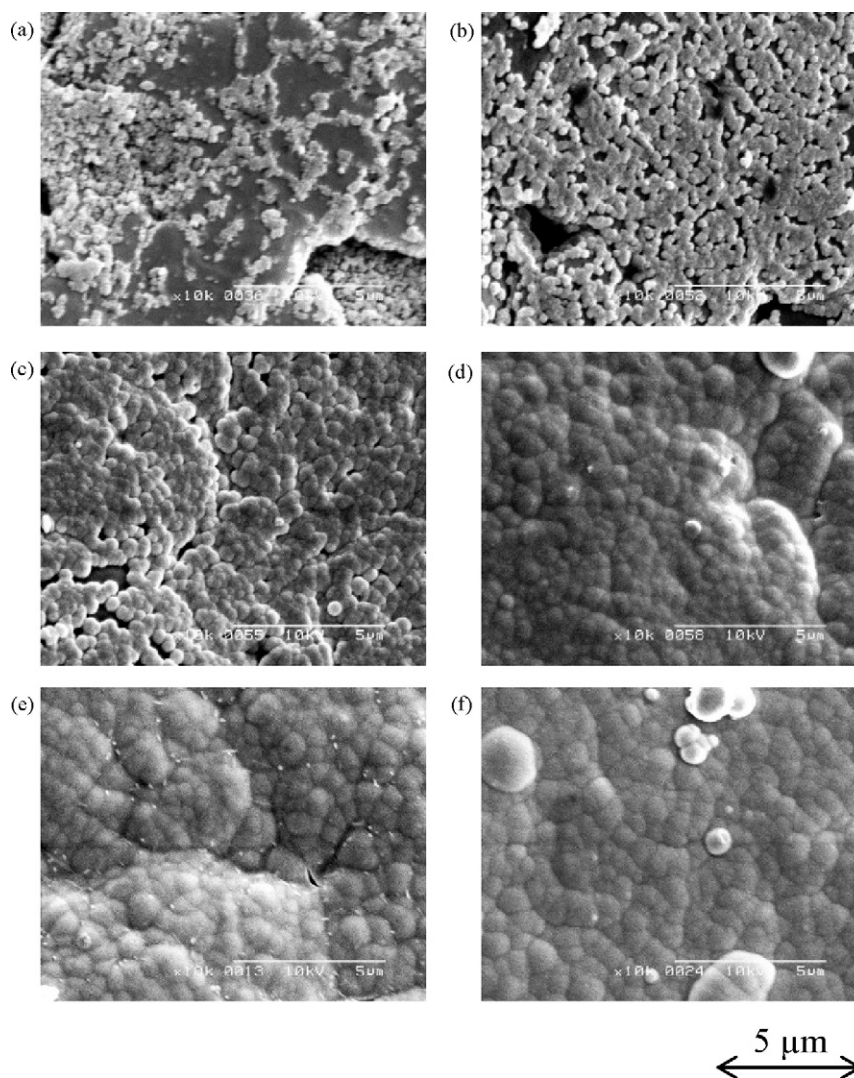
Fig. 5. SEM micrographs of Ni-PTFE with various Ni contents: (a) 0.53 wt%, (b) 1.29 wt%, (c) 8 wt%, (d) 4.69 wt%, (e) 6.12 wt%, and (f) 8.01 wt%.

material surface. This boundary might be attributable to the non-uniform deposition of Pd on the PTFE surface. Then Ni deposition occurred and covered the PTFE particle, as portrayed in Fig. 5(b)–(f). The dot-like Ni plated on the PTFE surface grew as islands in the first plating stage. Subsequently, the dots mutually connected to cover the PTFE surface with increasing Ni content. The PTFE surface was covered completely in cases of greater than 4.69 wt% of Ni content.

2.3. Characterization of the Ni deposited film by electroless plating

Using electroless plating method, Ni was plated onto the PTFE particle that had been hydrophilized using the surfactant and activated by Pd catalyst deposition. In fact, $C_{12}H_{25}$–O–$(C_2H_4O)_2$–H (BL-2) was selected as the surfactant to impart hydrophilicity to the PTFE particle surface. It gives high hydrophilicity to PTFE and has low effervescence during plating. Fig. 5 presents SEM micrographs of the sample with different Ni contents: Ni contents can be controlled by the deposition rate, which is the rate of adding the reducing agent.

Table 1 shows properties of Ni-plated PTFE. Fig. 6 shows the conductivity of Ni-plated PTFE with different Ni contents. Electric conductivity of the Ni-PTFE composite particle under 3 kg cm^{-2} increased with increasing Ni contents. The conductivity reached about $4 \times 10^2 \text{ S m}^{-1}$ when the Ni content was about 4.5 wt%. The conductivity remained steady for higher Ni contents. The SEM micrograph presented in Fig. 5
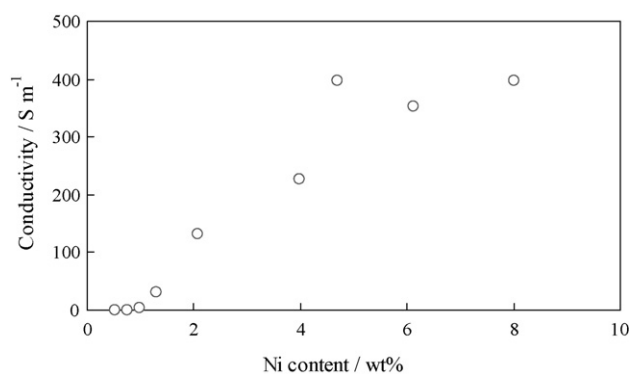

Fig. 6. Conductivity vs. Ni content.

Table 1
Characteristics of Ni-PTFE particles of various Ni contents

Ni content (wt%)	Conductivity (S m^{-1})	Density (g cm^{-3})	Total surface area (BET) (m^2 g^{-1})	Total pore volume (cm^3 g^{-1})	Average pore width (Å)
0.53	0	2.293	1.776	0.01374	309.4
0.76	4.2×10^{-1}	2.304			
0.99	3.7	2.305			
1.29	3.2×10	2.308	1.805	0.01545	342.3
2.08	1.3×10^2	2.336	1.550	0.01129	291.3
3.98	2.3×10^2	2.373			
4.69	4.0×10^2	2.389	1.698	0.01345	316.7
6.12	3.5×10^2	2.418	1.730	0.01539	355.7
8.01	4.0×10^2	2.450	4.908	0.00937	76.37

shows that the whole particle surface area was coated by Ni about 4.5 wt% of Ni contents.

The surface area (BET) shown in Table 1 and SEM micrographs in Fig. 5 show that the surface area and the pore size changed with the progress of Ni-plating. The surface area and the pore size apparently increased concomitant with the rugged area of the Ni-PTFE surface because the Ni-plating area extended from a dot to a rugged island along with the Ni-plating

Fig. 7. SEM micrographs of the crack in the Ni-PTFE film at Ni contents of (a) 6.12 wt% and (b) 8.01 wt%.

area's extension at the start of plating. Furthermore, because the rugged surface of Ni-plated PTFE became smooth, while the connecting Ni-plating area became an island to a continuum as the plating progressed, the surface area and the pore size decreased to about the level before plating. When it was plated further after the Ni-plating area became a continuum, the surface area increased because Ni was plated again on the continuum plating area with the rugged surface. Fig. 7 shows SEM micrographs of the crack of the Ni-plated PTFE. The crack was observed in case of Ni-plating greater than 6 wt%. The crack contributes to the increased surface area. The strain in the plated Ni film, which increases with increasing film thickness, might cause that crack formation.

2.4. Forming Ni-PTFE particle into the plate

Fig. 8 shows a photograph of the plate with a gas passage pattern formed by sintering after pressing of the Ni-PTFE. The respective sintering and pressing conditions were 350 °C and 10 kg cm^{-2}. Under these conditions, the PTFE was able to connect through the plated Ni film.

Fig. 9 portrays the pore diameter distribution, as measured using mercury porosimetry. Results show that the gas permeability increased when the pressing pressure decreased. The plate prepared at 13.2 kg cm^{-2} displayed peaks at 0.03, 1, 30, and 60 μm in Fig. 9(b), whereas that prepared at 500 kg cm^{-2} presented no peak, as shown in Fig. 9(a). Because the plate prepared at 500 kg cm^{-2} had no gas permeability, the profile in Fig. 9(a) can be referred as the baseline. The Ni film deposited on the PTFE particle contains many grain boundaries, as shown in Fig. 5. The peak in Fig. 9(b) at 0.03 μm might correspond to these grain boundaries, reflecting the roughness of the Ni film deposited on the PTFE particle. At least two kinds of pore were present in the Ni-PTFE plate prepared at 13.2 kg cm^{-2}, as shown in Fig. 10: pores along the boundary between two particles, and those among several particles. The former might correspond to the peak at 1 μm; the latter might correspond to the peaks at 30 and 60 μm. It is important to know how the gas permeates through these pores for controlling the pore distribution in the Ni-PTFE plate.

Fig. 11 shows the pore distributions in the plates produced by Ni-PTFE particles. Fine PTFE particles (M-12, 25 μm
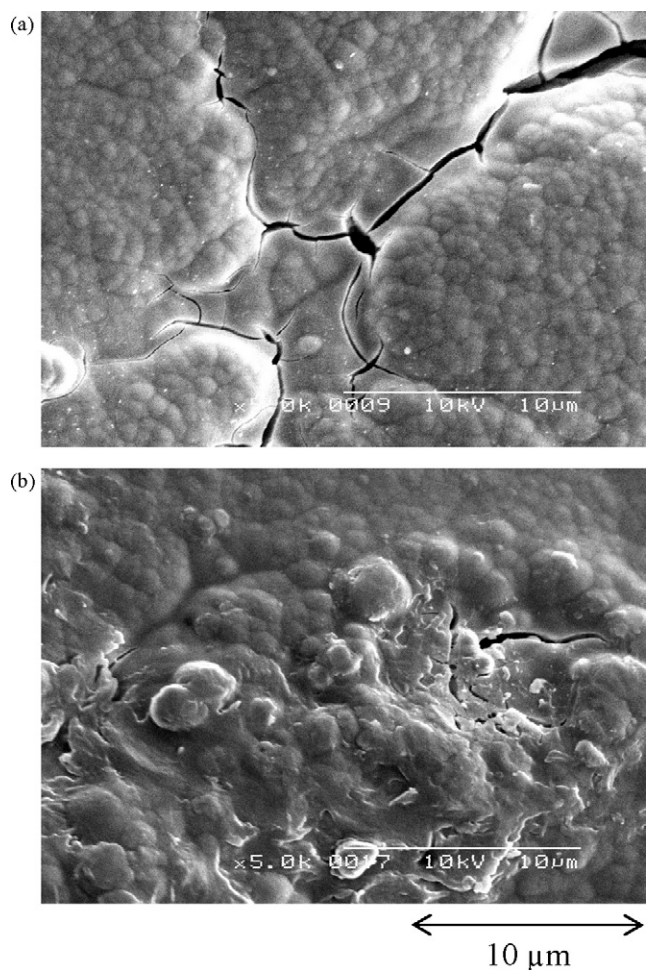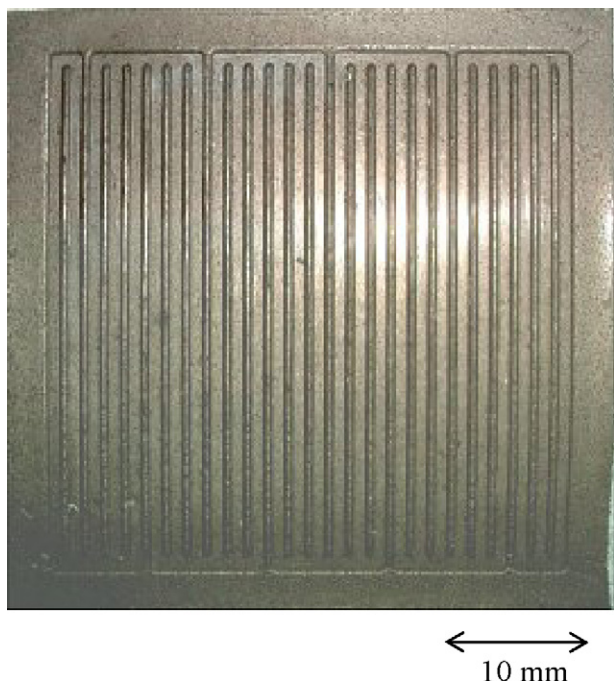
Fig. 8. Photograph of Ni-PTFE plate formed using Ni-PTFE fine powder under 500 kg cm^{-2}.

Fig. 10. SEM micrograph of Ni-PTFE plate cross section. (a) The pore along the boundary between two particles and (b) the pore among several particles.
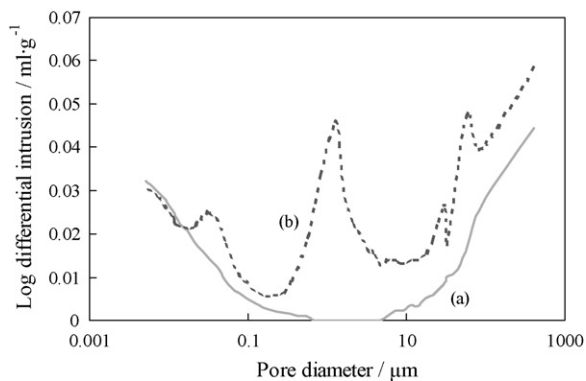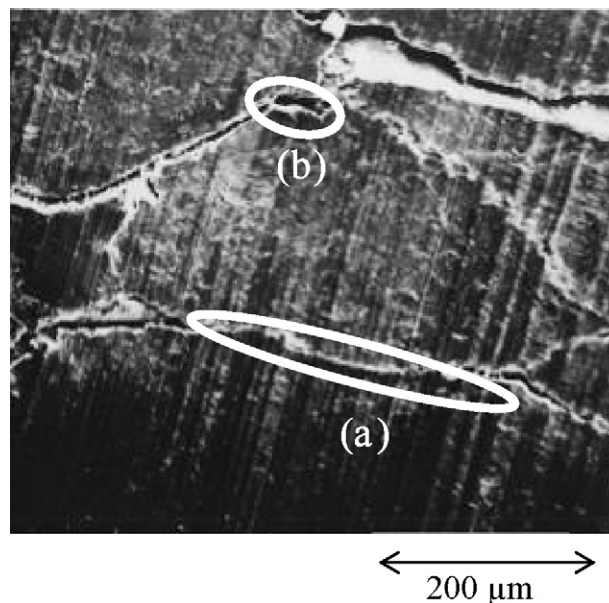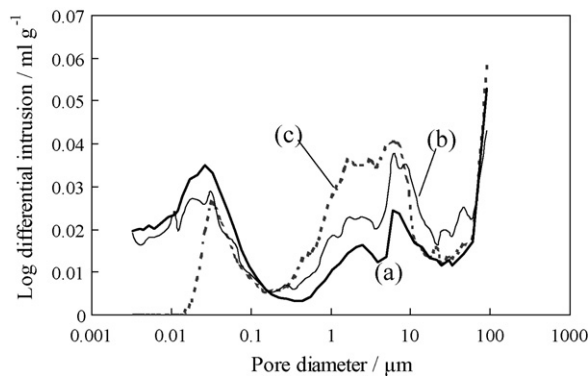

Fig. 9. Plot of pore diameter against log differential intrusion using mercury porosimetry for Ni-PTFE plate (M-393, Ø 500 μm). (a) 500 kg cm^{-2} and (b) 13.2 kg cm^{-2}.

Fig. 11. Plot of the pore diameter against log differential intrusion using mercury porosimetry for Ni-PTFE plate (M-12, Ø 25 μm). The solid line, dashed line, and dotted line, respectively, correspond to samples prepared under 1000 kg cm^{-2} (a), 500 kg cm^{-2} (b) and 300kg cm^{-2} (c).

Table 2
Conductivity of Ni-PTFE particles (M-12, Ø 25 μm) with various Ni contents

Ni content (wt%)	Conductivity (S m^{-1})
35	4.0×10
42	1.0×10^2
51	3.0×10^3
55	3.0×10^3

average particle size) were used in this case. A similar process to that for M-393 (500 μm average particle size) was used for electroless plating here. The profiles of the Ni-PTFE(M12) plates prepared under conditions of 1000, 500, and 300 kg cm^{-2} showed a difference at the peak at 1 μm. Increasing the pressure at pressing process, the number of pores of 1 μm decreased while pores with other diameters did not. The greater the pressure during the pressing process, the lower the gas permeability of the plate. It is therefore important to control the pore distribution, with emphasis on pores of 1 μm, to optimize the gas permeability of the Ni-PTFE plate.

Table 2 shows the conductivity of the Ni-PTFE(M-12) particle. In fact, 50 wt% of Ni content was necessary to cover the PTFE(M-12) particle with a 1 μm Ni film, while 5 wt% of Ni content was necessary to cover the PTFE(M-393) particle with a 1 μm Ni film. Consequently, the conductivity of Ni-

PTFE(M-12) was greater than that of Ni-PTFE(M-393) at the same Ni film thickness. Results demonstrate that thinner and more highly conductive plates can be prepared using Ni-PTFE(M-12).

3. Conclusion

In this study, PTFE particles were dispersed into water using nonionic hydrocarbon surfactant. Results show that the

hydrophilicity of surfactant-treated PTFE is influenced not only by the HLB value but also by the nature of the alkyl groups and the distribution of additional number of EO molecules to the nonionic surfactant. In addition, Ni was deposited by electroless deposition on surfactant-treated PTFE particles. Furthermore, Ni deposited partly on the PTFE surface because of the non-uniform deposition of Pd as a catalyst on the PTFE surface at the initial stage of the deposition. Electric conductivity of the Ni-PTFE composite particle of 3 kg cm^{-2} 2 increased with increasing Ni contents until the entire particle surface was covered with Ni (about 5 wt%). The Ni-PTFE plate was formed by 350 °C sintering after 10 kg cm^{-2} pressing of the Ni-PTFE particles. The PTFE was able to connect through the plated Ni film. The greater the pressure during the pressing process, the lower the gas permeability of the plate. It is important to control the pore distribution with specific emphasis on the pores of 1 μm to optimize the Ni-PTFE plate gas permeability. The conductivity of Ni-PTFE(M-12, 25 μm average particle size) was greater than that of Ni-PTFE(M-393, 500 μm average particle size) at the same thickness of the Ni film. Results show that a thinner and more highly conductive plate can be prepared using Ni-PTFE(M-12).

4. Experimental

As the plating core material to examine the plating conditions, we selected PTFE particles with 500 μm average diameter and 25 μm particle size (Polyflon PTFE M-393 and M-12; Daikin Industries, Ltd.), as depicted in Figs. 12 and 13. Many surfactants, hydrocarbon surfactants and homogeneous polyoxyethylene nonionic surfactants listed in Tables 3 and 4, were tested. As hydrophilic treatment, the PTFE particles were stirred in an aqueous solution of 2 wt% surfactant at 60 °C for 30 min and dried in a 70 °C air chamber after filtering and washing with ion-exchanged water. To retain sufficient hydrophilicity on the PTFE particles, about 100 ml water
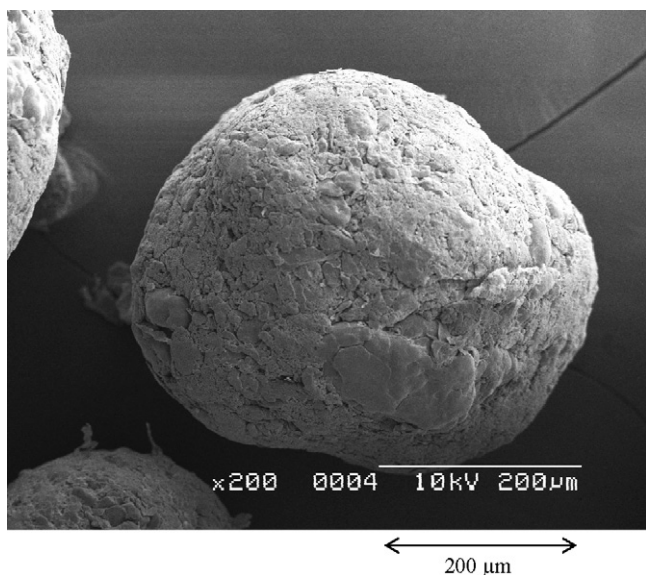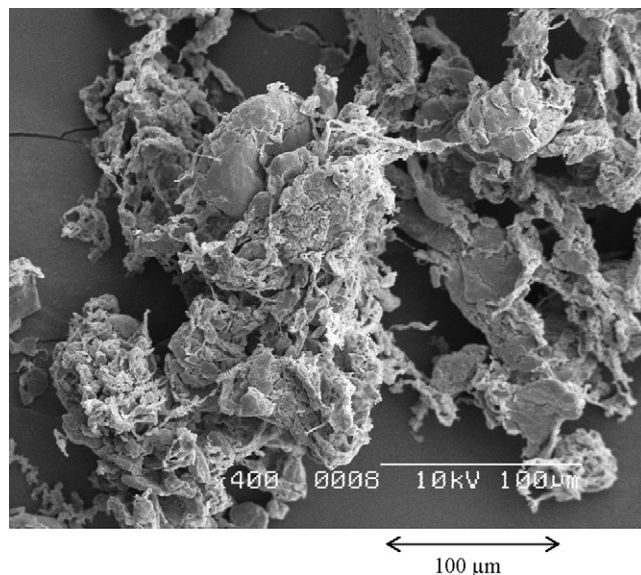

Fig. 13. SEM micrograph of Polyflon PTFE M-12.

was used for washing the 1 g PTFE particles. Hydrophilicity was evaluated from the contact angle of ion-exchanged water droplets (2-mm-diameter) on the surface of a plate produced by pressing 10 g of PTFE particles into 60 mm × 60 mm at 500 kg cm^{-2}. The contact angle was measured at 20 °C under relative humidity of 65% using a contact-angle meter (CA-S type 150; Kyowa Interface Science Co., Ltd.). The contact angle was measured 5 min after dropping a water droplet.

With respect to the sensitizing process for electroless metal plating, the PTFE particles were immersed in an aqueous

Table 3
Hydrocarbon surfactant

Ion type	Name	Structural formula
Cation	CA-2150	$[C_{12}H_{25}N(CH_3)_3]^+Cl^-$
Anion	SLS	$C_{12}H_{25}OSO_3Na$
	BL-2	$C_{12}H_{25}-O-(C_2H_4O)_2-H$
	BL-4.2	$C_{12}H_{25}-O-(C_2H_4O)_{4.2}-H$
	BL-9EX	$C_{12}H_{25}-O-(C_2H_4O)_9-H$
	BC-2	$C_{16}H_{33}-O-(C_2H_4O)_2-H$
Non-ion	BC-5.5	$C_{16}H_{33}-O-(C_2H_4O)_{5.5}-H$
	BS-2	$C_{18}H_{37}-O-(C_2H_4O)_2-H$
	BS-4	$C_{18}H_{37}-O-(C_2H_4O)_4-H$
	BO-2V	$C_{18}H_{35}-O-(C_2H_4O)_2-H$
	BO-7V	$C_{18}H_{35}-O-(C_2H_4O)_7-H$

*All fluorochemical surfactants were produced by Nikko Chemicals Co., Ltd.

Table 4
Homogeneous polyoxyethylene nonionic surfactant

Name	Structural formula
BL-1SY	$C_{12}H_{25}-O-(C_2H_4O)-H$
BL-2SY	$C_{12}H_{25}-O-(C_2H_4O)_2-H$
BL-3SY	$C_{12}H_{25}-O-(C_2H_4O)_3-H$
BL-4SY	$C_{12}H_{25}-O-(C_2H_4O)_4-H$
BL-5SY	$C_{12}H_{25}-O-(C_2H_4O)_5-H$
BL-6SY	$C_{12}H_{25}-O-(C_2H_4O)_6-H$

*All fluorochemical surfactants were produced by Nikko Chemicals Co., Ltd.

Fig. 12. SEM micrograph of Polyflon PTFE M-393.

solution of 2 wt% tin(II) chloride dihydrate (Kanto Chemical Co. Inc.) and 1 vol.% hydrochloric acid (12 M, Nacalai Tesque Inc.) for 10 min, followed by gentle rinsing with ion-exchanged water. For the activation process, sensitized PTFE particles were immersed in an aqueous solution of 0.1 wt% palladium(II) chloride (Mitsuwa Chemical Co., Ltd.) and 0.5 vol.% hydrochloric acid (12 M) for 2 min, followed by gentle rinsing with ion-exchanged water. The amount of Pd on the activated PTFE particles was measured using atomic absorption spectrometry (AAS, Z5000-300; Hitachi Ltd.) after dissolving Pd on the activated PTFE particles in nitric acid for AAS (Nacalai Tesque Inc.).

The electroless plating bath was prepared with a 20 g dm^{-3}-nickel(II) sulfate hexahydrate (Nacalai Tesque Inc.), 30 g dm^{-3}-tri-sodium citrate dihydrate (Nacalai Tesque Inc.), and sodium ammonium solution (Kanto Chemical Co. Inc.) as a pH adjuster. Then, sodium phosphinate monohydrate (Nacalai Tesque Inc.) was used as a reducing agent. The activated PTFE particles, 10 g, were put into the electroless bath of 1000 ml, which was controlled at 60 °C and pH 9.0. Finally, the substrate was rinsed carefully with ion-exchanged water and dried in a 70 °C air chamber after filtering. The conductivity of Ni-plated PTFE particles (Ni-PTFE) was measured using four-terminal DC method with a disk sample pressed at 3 kg cm^{-2}. The Ni-

PTFE surface was observed using SEM (S-2400; Hitachi Ltd.). The Ni amount was measured using AAS (Nacalai Tesque Inc.) from Ni film of Ni-PTFE in nitric acid. The electrode plate for the fuel cell was made by pressing the Ni-PTFE at 13.2 kg cm^{-2} in the size of 60 mm × 60 mm × 1 mm using a die with ditch pattern for gas flow; subsequently, it was sintered at 350 °C under 10%H$_2$–90%N$_2$ and atmospheric pressure for a hour. The gas permeability of the electrode plate was measured using automated mercury porosimetry (Auto Pore IV; Micrometrics Inc., Shimadzu Co.).

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