INTRODUCTION

Electrode of PEFC is fabricated by coating catalyst layer on carbon paper. Electrode structure depends on the coating method and catalyst paste viscosity, etc.

■ *How to coat catalyst layer on carbon paper*

- **Spray method**
- **Screen-printing method**

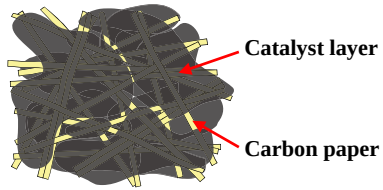


Fig. 1 Illustration of carbon paper coated by catalyst layer

The relationship between the electrode structure and impedance spectrum is investigated by electrochemical impedance spectroscopy (EIS).

■ *Electrode fabrication*

Amount and thickness of catalyst layer can be controlled precisely by using screen-printing method. In this study, hand screen-printing machine was used.

◆ ***Merits of hand screen-printing***

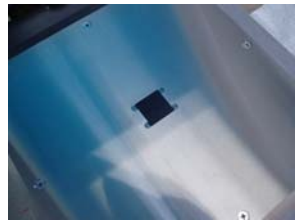
- **Easy to change printing conditions**
- **Short manufacturing time**

Fig. 2 Hand screen-printer designed for the fabrication of MEA (HP-320, NEWLONG Seimitsu Kogyo co., ltd.)

HAND SCREEN-PRINTING

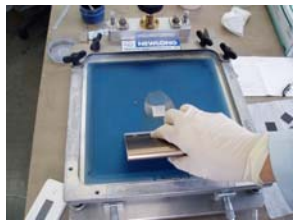
① Setting printing plate

② Setting carbon paper on the printing table

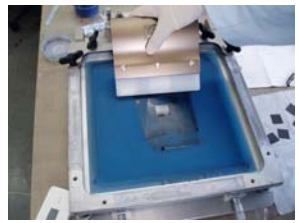


③ Putting catalyst paste on the printing plate

④ Filling catalyst paste in the pattern of the printing plate



⑤ Filling catalyst paste in the pattern of the printing plate



⑥ Printing catalyst paste on the carbon paper

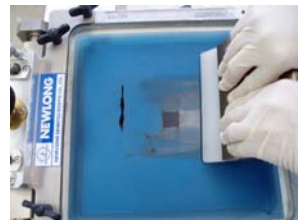
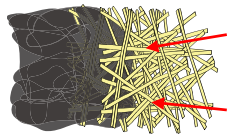


Fig. 3 Process of printing catalyst paste on carbon paper by using hand screen-printing machine

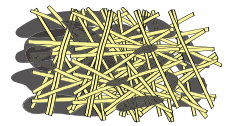
··· Fabricated two kinds of electrodes. (0.6 mg cm^{-2} Pt) ·····

Electrode-1



Most of catalyst layer is concentrated on the carbon paper

Electrode-2



Most of catalyst layer is distributed in the carbon paper

Fig. 4 Illustrations of electrode structure fabricated by hand screen-printing

FABRICATION OF MEA

Composition of the MEA

◆ **Gas diffusion layer :**
Carbon paper TGP-H-060 (Toray)

◆ **Electrolyte membrane :**
Nafion 117CS (DuPont)

◆ **Catalyst Paste**

- **Catalyst : Pt/C TEC10E50E (Tanaka Kikinzoku)**
- **Electrolyte : 5 wt% Nafion dispersion (DuPont)**
- **Solvent : Ethanol (Wako)**

Water repellent coating of carbon paper

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Screen-printing of catalyst paste

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Drying carbon paper (120 ºC, 15 min)

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Hot pressing carbon paper and electrolyte membrane (140 ºC, 2 MPa, 1 min)

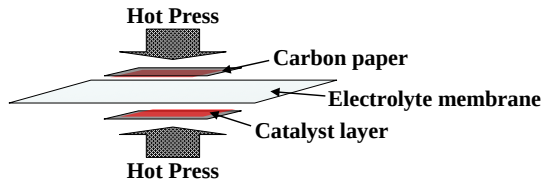


Fig. 5 Fabrication of the MEA by hot pressing carbon paper and electrolyte membrane

EXPERIMENTAL

■ *Operating test*

- Cell temperature : 72 ºC
- Humidifier temperature : 70 ºC
- Anode gas : H_2 1.0 L min^{-1}
- Cathode gas : O_2 1.0 L min^{-1}

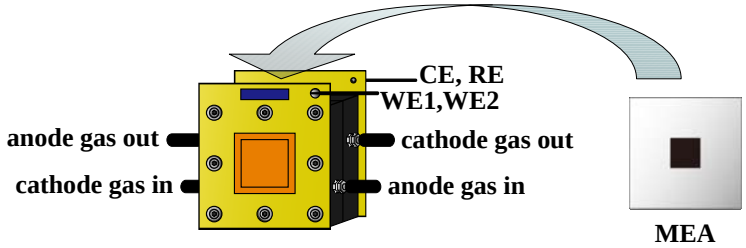


Fig. 6 Single cell for PEFC

■ *EIS measurement*

- Measurement voltage : 0.7 V
- AC potential amplitude : 10 mV
- Frequency range : 10 mHz – 1 kHz
- Integration time :
  - 1 times at 10 mHz – 1 Hz
  - 20 times at 1 Hz – 1 kHz

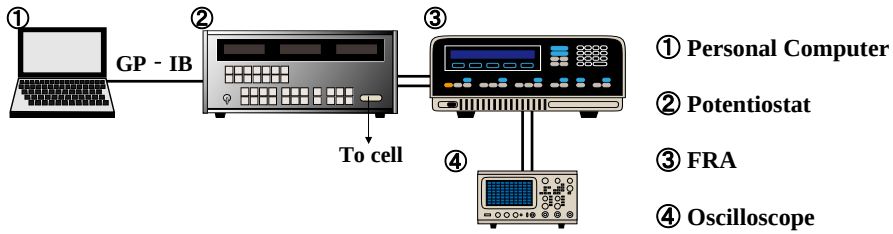
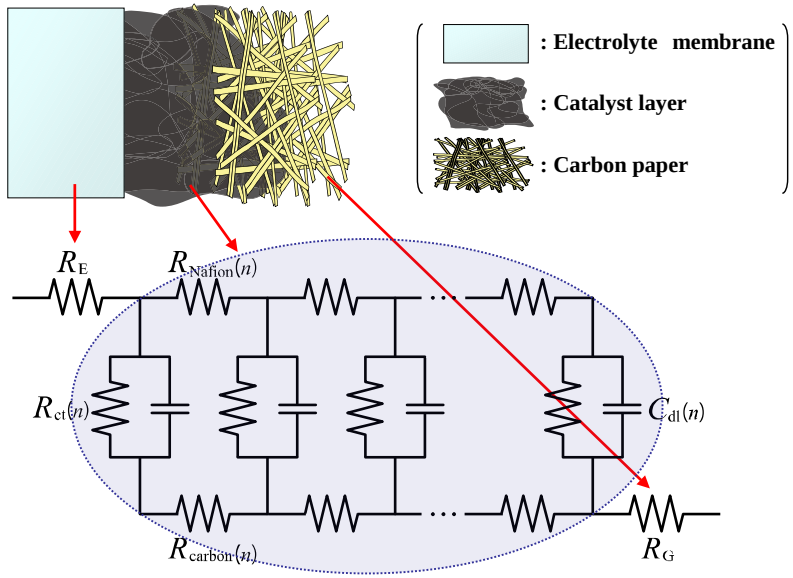


Fig. 7 Schematic diagram of the experimental apparatus

EQUIVALENT CIRCUIT

■ *Fitting*

In this study, distributed constant equivalent circuit shown in Fig. 8 was used in order to represent PEFC porous electrode. This equivalent circuit represents only cathode catalyst layer because the contribution from anode catalyst layer to total cell impedance can be negligible.[1)]



$R_{ct}(n)$ **: Charge transfer resistance (Ω)**
R_E **: Electrolyte membrane resistance (Ω)**
$R_{Nafion}(n)$ **: Cast Nafion resistance (Ω)**
R_G **: Gas diffusion layer resistance (Ω)**
$R_{Carbon}(n)$ **: Carbon resistance (Ω)**
$C_{dl}(n)$ **: Double layer capacitance (F)**

Fig. 8 Equivalent circuit for PEFC cathode catalyst layer

Reference

1) R. Makharia, M. F. Mathias, and D. R. Baker, J. Electrochem. Soc., 152, A970 (2005)

RESULTS

◆ *The MEA with Electrode-1*

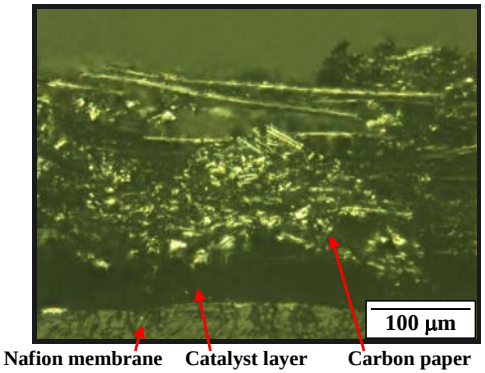


Fig. 9 Cross-sectional optical micrograph of *the MEA with Electrode-1*

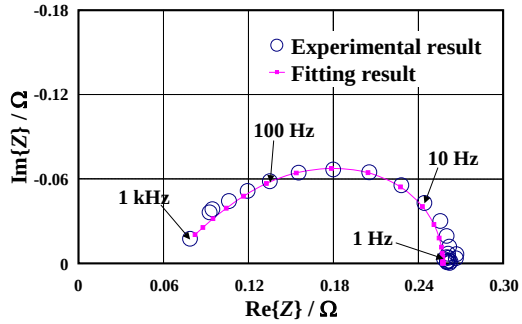


Fig. 10 Nyquist plots of impedance spectrum of *the MEA with Electrode-1*

Table 1 Fitting parameter for impedance spectrum of *the MEA with Electrode-1*

$R_{ct}(n)$	24.7
$R_{Nafion}(n)$	1.5×10^{-3}
$R_{Carbon}(n)$	7.0×10^{-7}
R_E	6.2×10^{-2}
R_G	0
C_{dl}	2.8×10^{-4}
Segment	300

◆ *The MEA with Electrode-2*

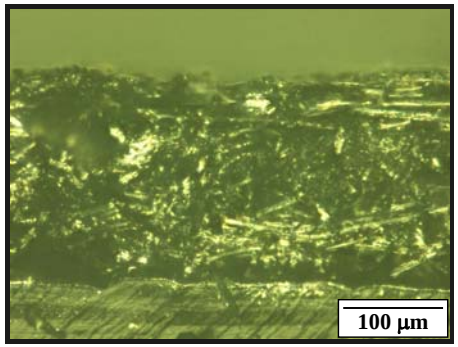


Fig. 11 Cross-sectional optical micrograph of *the MEA with Electrode-2*

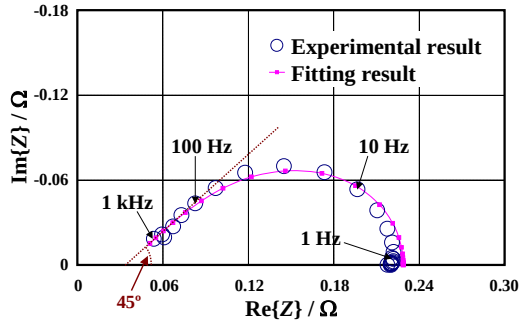


Fig. 12 Nyquist plots of impedance spectrum of *the MEA with Electrode-2*

Table 2 Fitting parameter for impedance spectrum of *the MEA with Electrode-2*

$R_{ct}(n)$	24.1
$R_{Nafion}(n)$	1.5×10^{-3}
$R_{Carbon}(n)$	7.0×10^{-7}
R_E	3.6×10^{-2}
R_G	0
C_{dl}	5.0×10^{-4}
Segment	300

- Diameter of the capacitive loop decreased when catalyst layer was distributed in the carbon paper.
- High-frequency limiting value decreased when catalyst layer was distributed in the carbon paper.
- Angle of straight line to real axis in high-frequency range was 45° when catalyst layer was distributed in the carbon paper.

DISCUSSION

◆ Diameter of the loop

Even though the amount of Pt is the same, the active area increases when catalyst layer is distributed in the carbon paper. Therefore, charge transfer resistance and diameter of the capacitive loop decreased for *the MEA with Electrode-2*.

◆ Electrolyte membrane resistance

The humidified gas is difficult to contact directly with the electrolyte membrane when the electrolyte membrane surface is coated with thick catalyst layer. (*the MEA with Electrode-1*) In contrast, the humidity gas becomes easy to contact directly with the electrolyte membrane when the catalyst layer surface is distributed in the carbon paper. Therefore, the electrolyte membrane resistance and high-frequency limiting value decreased for *the MEA with Electrode-2*.

◆ Angle of straight line to real axis in high-frequency range

Angle of straight line to real axis in high-frequency range is 45º for *the MEA with Electrode-2*. This is a typical behavior of electrode with distributed active area.

SUMMARY

- **Two kinds of electrodes with different structure were fabricated by using hand screen-printing machine.**
- **Relationship between electrode structure and impedance spectra was investigated.**