High performance of electrode with low Pt loading prepared by simplified direct screen printing process in PEM fuel cells

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Screen printing is one of the most popular methods used for the fabrication of a catalytic layer in the electrode of polymer electrolyte membrane fuel cells (PEMFCs), due to its convenience and adaptability, although many kinds of coating method have been applied such as hand brushing, screen printing [1], rolling [2] and spraying [3]. In the conventional screen printing process, Nafion membrane must be temporally changed to Na⁺ form to avoid swelling trouble during a hot press decal transferring because Na⁺ formed Nafion membrane is mechanically strong and stable at temperature ranges from 150 to 160 °C for hot pressing [4]. In the previous direct screen printing [5] process, the slurry was also applied to the membrane of Na⁺ form or tetra-butyl ammonium form to stabilize the catalytic layer enhancing the physical strength of the membrane. Anyhow, both coating processes are still subject to being complicated. In the study, we suggest an improved screen printing method which is a rather simple suppressing of the swelling trouble without an additive process and is also competitive, with very low Pt loadings in comparison to the previous methods. This method also makes it possible to omit the gasket loading step into the test cell.

For the MEA fabrication, we have used 20 wt%Pt/C (E-TEK), carbon paper (Sigracet GDL 10-H), Nafion 115^{TM} and 5 wt% Nafion solution (Dupont). The membrane, Nafion 115, was pre-treated by boiling in 3 wt% H₂O₂ and 0.5M H₂SO₄ solution [6]. And 20 wt% Pt/C (E-TEK), glycerin, Nafion solution and water were mixed for the preparation of a catalyst slurry. The mixed solution was uniformly dispersed by ultra-sonic method and stirring with heating for viscosity control.

For the gasket unified direct screen printing method, two polyester films (PP2910, 3M Inc.) with an open square window at the center were prepared and Nafion membrane was fixed between the transparency films. Then catalyst slurry was directly screen printed in the window followed by drying at 80 °C in an oven. The other side of the membrane was screen printed by the same process. In this method Nafion membrane was not required to be substituted to the Na⁺ form, and transparency film played a gasket role as well as support of the membrane. Swelling problem of the membrane could be suppressed by the adhesive strength between membrane and transparency film during the screen printing. Another simple process without gasket unified which omits Na⁺ substitution of membrane was performed. Well dried Nafion membrane was fixed to polyester films (PP2910, 3M Inc.) then it was screen printed. For catalyzing of the other side, membrane was hydrated in distilled water and dried again. This is also a simple method in which drying and hydrating is the key processe. The schematic diagram for the mentioned two processes is described in Fig. 1.

The catalyst layer was formed varying the number of screening while pressure, mesh size, scan speed and number of screening in screen printing method were fixed. Pt loadings of samples were analyzed by ICP AES method (Jobin Yvon ultima C).

Gas diffusion layer was combined by hot pressing at 90 atm for 1 min at 130 °C to the catalyzed face in the window on each side. Prior to hot pressing, the carbon paper was slightly brushed with carbon slurry to enhance adhesion between carbon paper and catalyzed membrane.

Fuel cells were operated with hydrogen and oxygen at 80 °C with 1 atm back pressure above atmospheric pressure by placing the MEA (membrane electrode assembly) in the single cell test fixture supplied by Electrochem Inc., USA. The cell was connected to fuel cell station to which humidified and heated hydrogen and oxygen gases were fed. Performance of the cell (I-V characteristics) was evaluated by Won-A-Tech (HPCS 1) system increasing the current to over 1000 mA/cm².

The ratio of area and weight of the Nafion membrane during the drying process at ambient atmosphere were measured and depicted in Fig. 2. The proportional changes of area and weight after 20 min were 17.5 and 16% respectively. The changes of membrane can be ascribed to the evaporation of water contained in the membrane. The adhesion of membrane to polyester film was best after about 10 min, although the adhesion cannot



1.0

0.8

0.6

0.4

Figure 1 MEA fabrication using two kind of renovated screen printing process: (a) Gasket unified and (b) without gasket unified.



Figure 2 The ratio of area and weight of Nafion membrane during the drying at ambient atmosphere (Initial area: 10.5 cm * 4.2 cm = 44.1 cm, initial weight: 1.072 g).



Cell Voltage(V) 0.2 0.0 200 400 600 1000 0 800 Current Density(mA/cm²) (a) 1.0 2 Layer without gasket 0.8 gasket unified Cell Voltage(V) 0.6 0.4 0.2 0.0 200 400 600 800 0 1000 Current Density(mA/cm²) (b)

1 Layer

without gasket

gasket unified

Figure 3 Cell performance and Pt loadings with the number of screen printing.

be quantified by any instrument unfortunately. Anyhow, enhancement of adhesive strength by drying the membrane plus using sticky polyester film as a supporter played a key role to prevent the swelling problem.

Figure 4 Comparison of cell performances between the gasket unified and the without gasket unified: (a) 1 layer and (b) 2 layers.

Using the simplified direct screen printing with gasket unified, catalytic layer was formed on the membrane. Cell performances and Pt loadings with the number of screen printing are described in Fig. 3. Pt loadings were almost doubled by the second screen printing. Difference of cell performance between two samples starts from about 0.025 V at low current density area then increased slowly to about 0.12 V at high current density area. The output voltages of 0.5 and 0.38 V could be obtained at 0.038 and 0.075 g/cm² of Pt loadings. It is very low Pt loadings.

On the other hand, a cell made without the gasket unified direct screen printing method was compared with the gasket unified (Fig. 4a and b). Cell performance was varied above current density of 400 mA/cm² for the same MEA. This discrepancy seems to be ascribed to the mass transport polarization related with gas consumption at high current density area. This means that the gasket unified can obstruct gas leakage better than that without gasket unified.

In summary, we have suggested simplified screen printing methods which are with or without gasket unified. The gasket unified showed better performances than the other method especially at high current area due to the blocking effect on the gas leakage during the operation. Cell performances by the suggested method were good enough. Especially the gasket unified performed output voltages of 0.5 and 0.38 V could be obtained at 0.038 and 0.075 g/cm² of Pt loadings. It is very low Pt loadings. New methods were competent to the previous methods in spite of eliminating the membrane treatment process to Na^+ form. Consequently, these methods give us more simplified and faster fabrication chances.

References

- 1. L. J. HOBSON, Y. NAKANO, H. OZU and S. HAYASE, J. Power Sources 104 (2002) 79.
- K. BOLWIN, E. GULZOW, D. BEVERS and W. SHUMBERGER, Solid State Ionics 77 (1995) 324.
- A. P. SAAB, F. H. GARZON and T. A. ZAWODZINSKI, J. Electrochem. Soc. 149 (2002) 1349.
- M. S. WILSON and S. GOTTESFELD, J. Appl. Electrochem. 22 (1992) 1.
- 5. M. S. WILSON, J. A. VALERIO and S. GOTTESFELD, *Electrochem. Acta* **40** (1995) 355.
- 6. A. T. ZAWODZINSKI, C. DEROUIN, JR., S. RADZINSKI, R. J. SHERMAN, W. T. SMITH, T. E. SPRINGER and S. Gottesfed, *J. Electrochem. Soc.* 140 (1993) 1041.

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