

Performance Improvement of Polymer Electrolyte Fuel Cell Electrode 고분자 연료전지 전극 성능향상에 대한 연구

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1. INTRODUCTION

Since fuel cells bring about the direct conversion of stored chemical energy to electrical energy without the intermediate generation of thermal energy, they are not limited by the Carnot cycle, unlike heat engines. Thus energy-conversion efficiencies for fuel cell systems (45-65%) are generally two times higher than those for heat engines [1]. Thus, there is a worldwide interest in the development and commercialization of polymer electrolyte membrane fuel cell (PEMFC) for vehicular and stationary applications. The success of the fuel cell is mainly related to the performance of the fuel cell electrode. Such high power densities are attained via better utilization of Pt crystallites in the reaction layer [2,3]. This enhanced performance can be achieved only when the reaction layer contains thin film of Pt particles on the front surface (membrane surface) as the bulk of the current generate close to the front surface of the electrode. It is reported that coating the catalyst slurry on the membrane leads to thinner reaction layer at the membrane surface which not only enhances the high oxygen reduction rate but also results in good contact between membrane and electrode, which is essential to minimize ohmic losses. This has resulted in the reduction of Pt loading from 4 mg to 0.4 mg per cm^2 with only a small loss in cell performance [4]. The robustness in the front surface catalysts is essential to minimize the coagulation of Pt particles when the fuel cells is subjected to long-term operation. This robustness of the catalyst structure depends on the manufacturing processes and also the organic solvents used to make the slurry.

In the present investigation, five different interfaces or surfaces were fabricated using different fabrication procedures. The results obtained using these procedures are discussed. The poison effect of CO was investigated at the anode interface. The

concentration of CO was maintained at 100ppm that is the concentration of CO normally present in reformat gas.

EXPERIMENTAL

When the organic solvents of different dielectric constant are mixed with Nafion solution [5], the resulted solution will be either one of the following form: solution, colloid, or precipitate. In the present investigation, solvents were selected with dielectric constants with greater than 10, except butyl acetate (5.01 at 20°C). However, isopropyl alcohol (18.1) was mixed with butyl acetate in such a manner that the dielectric constant of the resulted mixture was more than 10 and the slurry remains in solution.

Five different interfaces were prepared with different solvents and fabrication procedures using 30 wt% Pt/Ru (1:1) in Vulcan XC-72 (E-Tek, Inc, USA) for anode sides and 40wt% Pt/C (E-Tek, Inc, USA) for cathode sides. In all five electrode surfaces, the Pt loading of 0.7 mg/cm² were maintained for both anode and cathode surfaces. Nafion 115 from Dupont was used. The active electrode area was maintained at 50 cm² for all experiments.

Interface I was made using the catalyst slurry coated directly on the membrane. The slurry was made using 30wt%Pt/Ru for anode interface and 40wt% Pt/C for cathode interface. For both interfaces Nafion ionomers (5wt% solution), butyl acetate and isopropyl alcohol was added and sonicated for one hour.

For interface II, the chemical composition and preparation procedures were same as interface I except the 3-methyl butanol was used instead of butyl acetate and other solvents remains same. Interface II was coated directly on the membrane, like interface I.

Interface III was made using 5wt% Nafion ionomer, dimethyl sulphoxide isopropyl alcohol and 10wt% NaOH solution with 30wt% Pt/Ru for anode interface and 40wt% Pt/C for cathode interface. The membrane was ion-exchanged into Na⁺ before coating the slurry on the membrane. The coated membrane was baked at

150°C under vacuum for 3 hours and finally ion exchange to H⁺ form before use.

Interface IV was prepared same as interface III where ethylene glycol was used instead of dimethyl sulphoxide. The treatment procedure and the other ingredients were same as interface III.

Interface V was made using carbon cloth from M/S Textron Speciality Materials, USA, as electrode backing material. A gas diffusion layer was sprayed on to the carbon cloth, using Teflon as binding material, to a final concentration of 0.5mg C/cm². Then the Pt catalyst was coated on to the diffusion layer. The slurry was made using 30wt% Pt/Ru, 5wt% Nafion solution, glycerol, isopropyl alcohol and 10% NaOH solution for anode interface and 40wt% Pt/C with the same solvents for cathode interface. The electrode was baked at 150°C for 3 hours under vacuum. Before hot pressing, the electrode was converted to H⁺ form by treating with 0.5M H₂SO₄.

The experiments were carried out initially using H₂/O₂ and then H₂/air, subsequently the behavior of the electrode was studied using H₂ containing 100ppm CO. In all experiments, the polarization data were recorded at various current densities at different temperatures.

RESULTS AND DISCUSSION

Figure 1 shows the polarization curves for interface I using Nafion 115 with H₂/O₂, H₂/air, H₂+CO/O₂ and H₂+CO/air. Interface I was prepared by coating the catalyst slurry directly on the membrane. The maximum current density of 1100 mA/cm² at 0.5V at 80°C was obtained using H₂ and O₂ and 640 mA/cm² at 0.5V at 80°C was obtained using H₂ and air at ambient pressure.

The behaviour of the electrode was investigated by introducing a small amount of CO (100ppm) in H₂ compartment by using oxygen in cathode compartment. There was not much decay in cell performance when oxygen was used in cathode compartment, however decay in cell performance was observed when air was used instead of oxygen. This decay in cell performance was due to the partial pressure of oxygen in air at the cathode side and poisoning of the electrode surface by CO on the

anode side [8,9].

The polarization curves for interface I using Nafion 115 with H₂/O₂, H₂/air, H₂+CO/O₂ and H₂+CO/air are demonstrated in Figure 2.. Interface II was prepared by coating the slurry directly on the membrane. It gives high performance of 1200, 800 mA/cm² at 0.5 and 0.6V at 80^oC and ambient pressure using cathode O₂. With air, it gives a current density of 700, 500 mA/cm² at 0.5 and 0.6V at 80^oC and ambient pressure, shown in Fig.11. Experiments were also conducted with H₂ containing CO as anode gas and oxygen or air as cathode gases. The variations of cell performance using pure hydrogen and hydrogen mixed with CO was about 2% for oxygen and 7% for air.

Figure 3 shows the polarization data for interface III coated on the membrane directly. It gives current density of 720 mA/cm² for H₂/O₂ and 480 mA/cm² for H₂/air at 0.5V at 80^oC and ambient pressure. The performance varies only 2% using H₂/O₂ from 60 to 80^oC and 8% for H₂/air.

Figure 4 shows the polarization curves for interface IV at 80^oC using H₂, H₂+CO with O₂ and air prepared by coating the membrane. The fabrication procedure of Interface IV was similar to interface III. A current density of 800 mA/cm² for H₂/O₂ and 520 mA/cm² for H₂/air was obtained at 0.5V at 80^oC and ambient pressure.

The polarization data for interface V using H₂/O₂, H₂/air, H₂+CO/O₂, H₂+CO/air at 80^oC and ambient pressure are shown in Figure 5. Interface V was prepared by hot pressing the electrode with membrane. This interface was different from the other interfaces where the slurry was coated directly on the carbon cloth substrate material. The maximum current density of 800 mA/cm² at 0.5V at 80^oC using H₂/O₂ and 480 mA/cm² at 0.5V at 80^oC using H₂/air at ambient pressure was observed. When CO was mixed with anode compartment, a slight decay in cell performance (about 3%) similar to other interfaces I-IV was observed using O₂ cathode. When air was used, decay in cell performance was 9%.

CONCLUSIONS

Interface II which was prepared by coating the catalyst layer directly on Nafion membrane and using 3-methyl butanol as a solvent gives higher performance of 1200 mA/cm² at 0.5 V and 800 mA/cm² at 0.6V than any other interface due to the minimization of agglomeration of Pt particles.

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FIGURES

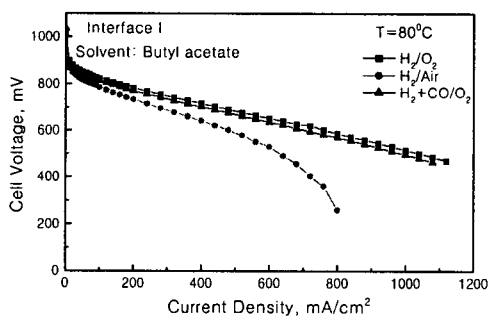


Fig. 1. Polarization curves for interface I using Nafion 115 with H₂/O₂, H₂/air and H₂+CO/O₂

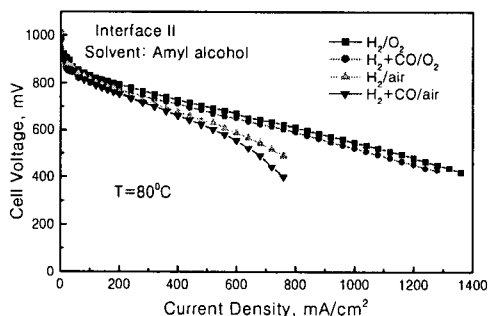


Fig. 2. Polarization curves for interface II using Nafion 115 with H₂/O₂, H₂/air, H₂+CO/O₂ and H₂+CO/air

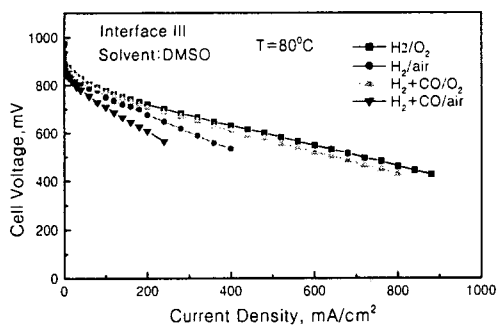


Fig. 3. Polarization curves for interface III using Nafion 115 with H_2/O_2 , H_2/air , H_2+CO/O_2 and H_2+CO/air

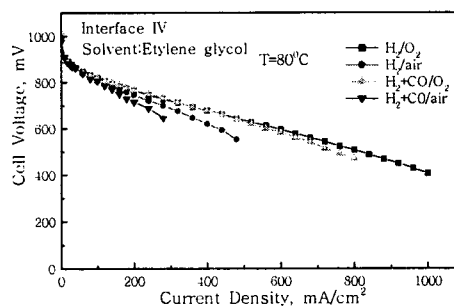


Fig. 4. Polarization curves for interface IV using Nafion 115 with H_2/O_2 , H_2/air , H_2+CO/O_2 and H_2+CO/air

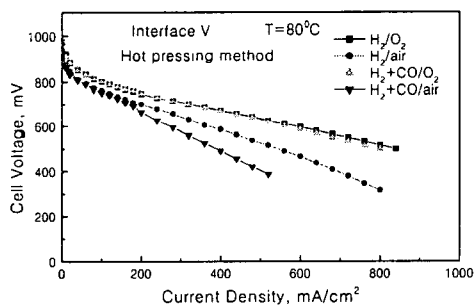


Fig. 5. Polarization curves for interface IV using Nafion 115 with H_2/O_2 , H_2/air , H_2+CO/O_2 and H_2+CO/air