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An investigation into multi-layered coatings on bipolar plates for a PEM (proton exchange membrane) fuel cell application

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Stainless steel 304 and 316 plates were deposited with multi-layered coatings of titanium films $(0.1 \ \mu\text{m})$ and gold films $(1-2 \ \mu\text{m})$ by an electron beam evaporation method. XRD patterns of the stainless steel plates modified with the multi-layered coatings showed the crystalline phases of the external gold film and the stainless steel substrate. Surface microstructural morphologies of the stainless steel bipolar plates modified with multi-layered coatings were observed by atomic force microscope (AFM) and field emission-scanning electron microscope (FE-SEM) images. The external gold films formed on the stainless steel plates showed the microstructure of grains of about 100 nm diameter. The grain size of the external surface of the stainless steel plates increased with the gold film thickness. The electrical resistivity and water contact angle of the stainless steel bipolar plates covered with multi-layered coatings were examined as a function of the thickness of the external gold film.

Key words: Proton exchange membrane fuel cell (PEMFC), Bipolar plates, Gold film, Titanium film, Resistivity.

Introduction

In recent years, escalating oil prices and serious global warming have become worldwide problems. In order to decrease greenhouse gases such as carbon dioxide, utilization of fuel cells in transportation and stationary power are essential. Proton exchange membrane fuel cells (PEMFCs) are the most promising power sources in the near future for transportation applications [1]. Hydrogen-powered fuel cell vehicle technologies have been developed by many researches and efforts are being made towards their commercialization. Increasing attention is being paid to the developments of proton exchange membrane fuel cells, because they operate at low temperatures and show rapid start-up, high efficiency and near-zero emissions [2, 3]. A key component of PEMFCs is the bipolar plate, which serves to electrically connect the electrodes of the stacks to obtain a useful voltage [4, 5]. In current technologies, the most costly component in PEMFC is the bipolar plate that is currently machined from bulk graphite or hot-molded using a composite carbon and resin [6]. The present PEM fuel cells utilize machined, very thick and heavy graphite blocks (as a bipolar plate). For commercialization of PEMFCs, it is necessary to reduce the manufacturing cost and weight of the fuel cell components [7, 8]. As alternative bipolar plate materials, carbon composites and metal plates have been studied for

PEMFCs [7, 9]. The development of metallic bipolar plates using stainless steels requires the prevention of the degradation, caused by corrosion of the bipolar plates in acidic conditions. Protective and conductive layers have been considered for the maintenance of electrical properties and stability of metallic bipolar plates in PEMFCs [9-13].

In this study, stainless steel 304 and 316 plate were selected as metallic substrates for bipolar plates and deposited with multi-layered coatings of titanium and gold films. The deposition process to form the titanium and gold films on the stainless steel bipolar plates was carried out by an electron beam evaporation method. The crystal phases of the metallic plates modified with the multi-layered coatings and bare stainless steels were identified from X-ray diffraction (XRD) patterns. The electrical resistivity of the stainless steel plates coated was measured. The surface morphologies of the stainless steel plates modified and bare stainless steels were observed by FE-SEM, AFM. Contact angles of the coated-stainless steel bipolar plates and the bare stainless steel plates were measured.

Experimental Procedure

Commercial stainless steel 304 and 316 plates were selected as the substrates for metallic bipolar plates for a proton exchange membrane fuel cell (PEMFC) application. The surfaces of the stainless steel plates were cleaned with isopropylalcohol. The surfaces of the stainless steel 304 and 316 plates were deposited with an external gold film (1-2 μ m) and a titanium film (0.1 μ m). The deposition of the Au film 1um thick was carried out for 30 minutes. After the deposition procedure of the conductive films of gold

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and titanium, the electrical resistivity of the stainless steel plates was evaluated. The formation of the multi-layered coatings on stainless steel plates was conducted by an Ebeam method (Electron Beam Evaporator, World Science Co., Korea). The crystalline phases of the multi-layered coatings coated on the stainless steel plates were identified by an X-ray diffractometer (Xpert PRO, PAnalitical, The Netherlands). The electrical resistivity and i-v characteristics of the stainless steel plates coated with the conductive films and the bare stainless steel plates were measured by an i-v source meter (Keithley, USA). The surface microstructural morphologies of the multi-layered coatings coated on stainless steel plates was observed by a scanning probe microscope (XE-200, PSIA corp. Korea) and a field emission scanning electron microscope (FE-SEM, JSM 6700F, JEOL, Japan). The water contact angles of the metallic bipolar plates modified using multi-layered coatings were measured by a contact angle meter (Digidrop, GBX, France).

Results and Discussion

XRD patterns of multi-layered coatings deposited on stainless steel bipolar plates

Fig. 1 shows XRD patterns of multi-layered coatings of external gold and titanium films deposited on stainless steel bipolar plates and the as-received stainless steel plate. The metallic bipolar plates coated with the multilayered coatings showed a γ -Fe phase pattern of the stainless steel. The XRD patterns of the modified bipolar plates did not showed obvious dependence on the different kinds of stainless steel. The multi-layered coatings formed on the stainless steel plates showed four strong peaks (38.32°, 43.81°, 64.62°, 77.58°) corresponding to the (111), (200), (220) and (311) reflections of the Au phase (the external film, thickness: 1-2 µm) in the XRD patterns. The peaks from the Ti phase, deposited as the interlayer (thickness: 0.1 µm) between the external gold film and stainless steel substrate, were not detected in these XRD patterns.

Surface morphology of the metal bipolar plates modified with multi-layered coatings

Fig. 2 shows AFM surface morphologies of the metallic bipolar plates modified with the titanium interlayer (0.1 mm) and the external gold film (1-2 mm). Fig. 3 shows FE-SEM images of the stainless steel plates deposited with the multi-layered coatings of the external gold film and titanium film. Surface microstructure has been observed mainly near in the center region of the modified stainless steel plates. Surface morphologies with a grain size of approximately 100 nm were shown in the external gold film on the modified stainless steel plates. With an increase of the thickness of the gold film to 2 mm, the grain size of the surface grains, observed in FE-SEM images, increased compared to that of the steel plate with a 1 mm Au film. Probably, the deposition process of the external surface film for 60 minutes induced the grain growth of Au grains.



Fig. 1. (a). XRD patterns of the stainless steel 304 plates deposited with Ti and Au.

Current-voltage characteristics of the metal bipolar plates modified with multi-layered coatings

Fig. 4 shows the current-voltage characteristics of the stainless steel plates deposited with the multi-layered coatings and the bare-SUS 304, 316 plates. A prominent decline of the i-v curve slope of the stainless steel plates, modified with the multi-layered coatings composed of the external gold film and titanium film, are shown in Fig. 4(a), (b). With the increase of the thickness of the external gold film from 1 µm to 2 µm, the stainless steel plates modified with the multi-layered coatings showed a lower electrical resistivity. The modified stainless steel 304 and 316 plates showed a similar i-v curve slope change in the current-voltage relationship. The modified stainless steel 304 plate with 2 μ m Au film and 0.1 μ m Ti film showed lowest electrical resistance (2.61 ohm) in the resistance-current relationship. The electrical resistance of the modified steel 304 and 316 plates showed a tendency to decrease by increasing the thickness the external gold layer from 1 µm to 2 µm. It was inferred that the electrical resistance obtained from the surface regions of the modified plates was affected by the surface oxide films created on bare stainless steel. To reduce



Fig. 2. AFM images of the stainless steel 304, 316 plates deposited with Ti and Au.



Fig. 3. FE-SEM images of the stainless steel 304, 316 plates deposited with Ti and Au ($1-2 \mu mAu / 0.1 \mu m$ Ti coating).



Fig. 4. (a) I-V characteristics of the stainless steel 304, 316 plates deposited with Ti and Au

the electrical resistance, the surface oxide layers (with chemical composition such as Fe-Cr-Ni-O) have to be removed properly through surface treatment prior to form conductive coating.

Contact angles of the metal bipolar plates modified with multi-layered coatings

Fig. 5 shows the contact angles of the bare stainless steel plates and the bipolar plates coated with the multilayered coatings of an external gold film and titanium film. The bare-SUS 304, 316 plates showed approximately 60 degree contact angles and the stainless steel bipolar plates modified with multi-layered coatings showed very high contact angles for water over 90 degree in average values. Especially, the stainless steel 304 plates coated with the multi-layered coatings of external gold film (1-2 mm) and titanium film represented prominent hydrophobic behavior with average contact angles of 98.5 degree and 99 degree, respectively. A high water contact angle of the bipolar plates in a PEM fuel cell is required for the effective drainage or circulation of water created during cell operation. In this study, the deposition of the multi-layered films of gold and titanium was able to increase the water contact angles of stainless steel plates.



Fig. 5. Contact angles of stainless steel 304, 316 plates deposited with Ti and Au. TA-110 (1 μ m Au/0.1 μ m Ti), TA-120 (2 μ m Au/0.1 μ m Ti).

Conclusions

Multi-layered coatings of a titanium interlayer (with a thickness of 0.1 µm) and external gold film (with a thickness of 1-2 µm) were formed on surface of stainless steel 304 and 316 plates through an electron beam evaporation process. The external gold surface morphologies of the modified metal bipolar plates exhibited grains of about 100 nm diameter. The metallic bipolar plates coated with the multi-layered coatings showed the γ -Fe phase pattern of stainless steel as well as strong peaks (38.32°, 43.81°, 64.62°, 77.58°) corresponding to the (111), (200), (220) and (311) reflections of the Au phase (the external film) in XRD patterns. The electrical resistance of the stainless steel plates, modified with multi-layered coatings of a Ti film and a Au film, decreased with an increase of the external film thickness from 1 µm to 2 µm. The stainless steel bipolar plates modified showed high water contact angles in the range of 90 degree to 99 degree (average values).

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