

Stainless steel bipolar plates for PEMFC application

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ABSTRACT: The CrN (Cr₂N)-chromium-ITO (indium-tin oxide) system and the gold-titanium system were deposited on the stainless steel 316 and 304 plates by a sputtering method and an E-beam method, respectively. The XRD patterns of the stainless steel plates deposited showed the crystalline phase of a typical indium-tin oxide, metallic phases such as chromium, gold and the metal substrate as well as external chromium nitride films. The nitride films were composed of two metal nitride phases of CrN and Cr₂N compounds. Surface morphologies of the stainless steel bipolar plates deposited were observed by AFM and FE-SEM. The stainless steel plates deposited with CrN (Cr₂N)-chromium-ITO system represented the surface microstructural morphology of the fine columnar grains of 10nm diameter and 60nm length. The external gold films formed on the stainless steel plates showed microstructure of grains of about 100nm diameter. The grain size of the external surface of the stainless steel plates deposited with gold-titanium system increased with the gold film thickness. The electrical resistance and water contact angle of the stainless steel bipolar plates covered with multi-layered coatings were examined with the thickness of ITO film or external gold film.

Keywords: Proton exchange membrane fuel cell (PEMFC), Bipolar plates, Resistance

1. INTRODUCTION

Proton exchange membrane fuel cells (PEMFCs) are the most promising power source in the near future for transportation application. [1] In recent, a new generation of vehicle programs was focused on the development of fuel cell vehicle technologies. [2. 3] Such hydrogen-powered fuel cell vehicle technologies require fuel cells with high power density, and a rapid response property to loads as well as the hydrogen-supply infrastructure. PEMFC system shows relatively low working temperature, a high efficiency, a high power density and a rather rapid response to the load variation. In present, the fabrication and commercialization of PEMFC stack and

its major components are very important for the utilization in several power generation systems, especially new generation of vehicle programs and such hydrogen-powered fuel cell vehicles. [4-6] PEM fuel cell technologies are closely related to polymer electrolyte membrane, metallic catalyst, balance of plant (BOP) and bipolar plates. Above all, the cost and weight of manifold bipolar plates in PEMFC are the chief obstacles to commercialization. The present bipolar plates for PEMFCs are the machined, very thick and heavy graphite blocks. [6] The manifold graphite bipolar plates in the present PEMFC system would occupy large dimensions, weight fraction over 70-80% as well as fabrication cost ratio of 40-45%. [7-8]

A key component of PEMFC is the bipolar plate, which serves to electrically connect the electrode of the stacks to obtain a useful voltage. [5] In the 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. The present PEM fuel cells utilize the machined, very thick and heavy graphite blocks (bipolar plate). For commercialization of PEMFC, it is required to reduce the manufacturing cost and weight of the fuel cell components. [7] As alternative bipolar plate materials, carbon composites and metal plates have been studied for the PEMFC. [8] The development of metallic bipolar plates such as 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 PEMFC. [9-12]

In this study, the stainless steel 304 and 316 plates were selected as the metallic bipolar plates. For the surface modification of stainless steel 304 and 316 plate, the CrN (Cr₂N)-chromium-ITO (indium-tin oxide) system and the gold-titanium system were introduced into the multi-layered film deposition by magnetron sputtering system and electron-beam evaporation system. The crystal phases of the metallic plates modified with the multi-layered coating systems and bare stainless steels were identified from XRD patterns. The electrical resistance of the stainless steel plates coated and bare stainless steels were measured. Surface morphologies of the stainless steel plates modified and bare stainless steels were observed by field emission secondary electron microscopy (FE-SEM) and atomic force microscopy (AFM). The water contact angles of the coated-stainless steel bipolar plates and the bare stainless steel plates were measured.

2. Experimental procedure

Commercial stainless steel 304 and 316 plates were selected as the substrates of bipolar plates for proton exchange membrane fuel cell (PEMFC) application. The chromium-ITO (indium-tin oxide) system and the gold-titanium system were deposited on stainless steel 304 and 316 plates by a sputtering method (Vacuum Science Co. Korea) and an E-beam method (Electron Beam Evaporator, World Science Co., Korea). External metallic film (100nm) and conductive interlayer (200-600nm) were coated to the surface of stainless steel 316, 304 plates. In other deposition procedure, the surface of the stainless steel 304, 316 plates was deposited with the external gold film (1-2μm) and titanium film (0.1μm). The crystalline phases of the multi-layered coatings coated on the stainless steel plates were identified by an X-ray diffractometer (Xpert PRO, PANalytical, The Netherlands). The electrical resistance and I-V characteristics of the stainless steel plates coated with multi-layered coatings 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 microscopy (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) and contact angle measuring system (Easydrop, KRÜSS, Germany).

3. Results and discussion

3.1 X-ray diffraction patterns of stainless steel plates deposited with multi-layered coatings

Fig. 1 shows the XRD patterns of the stainless steel bipolar plates modified with metallic chromium film and ITO interlayer.

The metallic plates deposited with chromium-ITO system showed a typical γ -Fe phase pattern of stainless steel and indium-tin oxide pattern and metallic chromium pattern. Also, the multi-film coated bipolar plates indicated the presence of crystalline metal nitride phases. It was inferred that chromium nitride compounds such as CrN or Cr₂N on the external surface were formed by the heat treatment procedure for the chromium coating on the stainless steel plates. The presence of the nitride phase on the coated stainless steel bipolar plates would have some influence on the water contact behavior and electrical resistance of the surface region. The XRD patterns of metallic film coated-steel plates without the conductive film showed typical a γ -Fe phase of the steel and a metal phase. The stainless steel bipolar plates coated with a metallic film and conductive interlayer showed the typical γ -Fe phase of steel, a weak metal nitride phase and a metal phase.

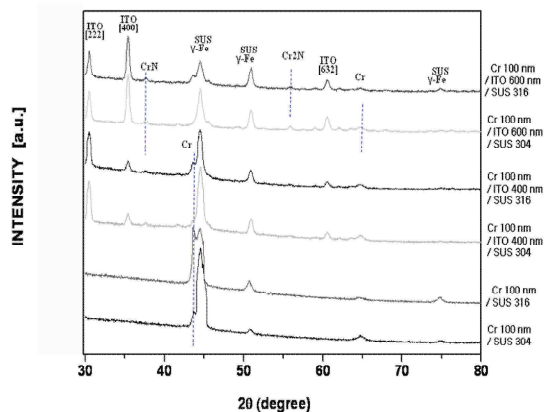


Figure 1: X-ray patterns of chromium film and ITO film on stainless steel 304 and 316 plates

Fig. 2 shows the XRD patterns of the multi-layered coatings of the external gold film and titanium film deposited on stainless steel bipolar plates. The metallic bipolar plates coated with the multi-layered coatings showed a γ -Fe phase pattern of stainless steel. The multi-layered coatings formed on the stainless steel plates showed four strong peaks corresponding to the (111), (200),

(220) and (311) reflection of the Au phase (thickness: 1-2μm) in XRD patterns.

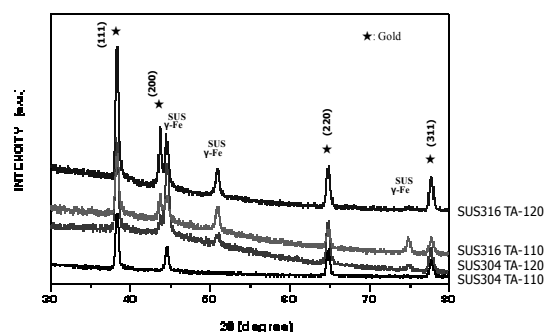


Figure 2: X-ray patterns of gold film and titanium film on stainless steel 304 and 316 plates. TA-110 (1μm Au / 0.1μm Ti), TA-120 (2μm Au / 0.1μm Ti)

3.2 Surface morphology of stainless steel plates deposited with multi-layered coatings

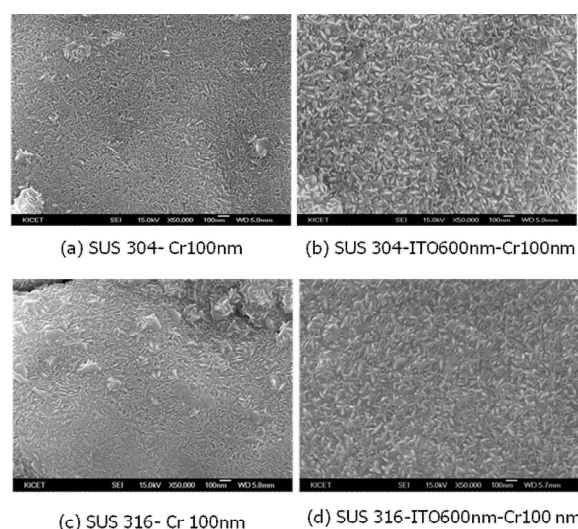


Figure 3: FE-SEM images of chromium film and ITO film coated-SUS 304 and 316 plates.

Fig. 3 shows FE-SEM images of the stainless steel bipolar plates modified with the CrN (Cr₂N)-chromium-ITO system. The external metallic film formed on the stainless steel plates demonstrated the microstructural morphology of nano-scale fine bars or columnar-like shaped grains regardless of the presence of the intermediate ITO film. The fine columnar grains of the external surface show a

uniform shape of approximately 10nm diameter and 60nm length. These crystalline grains showed some size variation according to the conditions of the film formation and interlayer thickness. It seemed that the size of the unusual crystalline grains was dependent upon the thickness of the conductive interlayer.

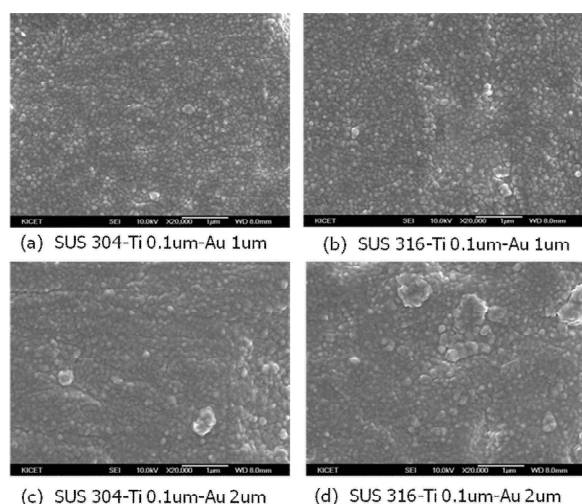


Figure 4: FE-SEM images of the stainless steel 304, 316 plates deposited with Ti and Au. (1-2um Au / 0.1um Ti film)

Fig. 4 shows FE-SEM images of the stainless steel plates deposited with the multi-layered coatings of the external gold film and titanium film. Surface morphologies with grain size of approximately 100nm were shown in the external gold film on the stainless steel plates modified. As increasing thickness of the gold film to 2um, the grain size of the surface grains, observed in FE-SEM images, increased compared to that of morphology of steel plate with 1um Au film. Probably, the deposition process of external surface film for 60 minutes induced the grain growth of Au grains.

3.3 I-V curve and electrical resistance of stainless steel plates modified with multi-layered coatings

Fig. 5 shows the current-voltage characteristics of the bare-SUS 304, 316

plates and the stainless steel plates deposited with CrN (Cr_2N)-chromium-ITO system. The I-V curve of the multi-film coated bipolar plates showed rather a rapid change in increment compared to the bare-SUS plates. The bare stainless steel plates and the stainless steel plates coated with only the metallic film showed rather similar features in the current-voltage variation. The stainless steel 304 and 316 bipolar plates, which were modified with only the metallic chromium film, showed relatively low resistance of about 5 Ohm and 6 Ohm compared to the bare-SUS plates (Table 1).

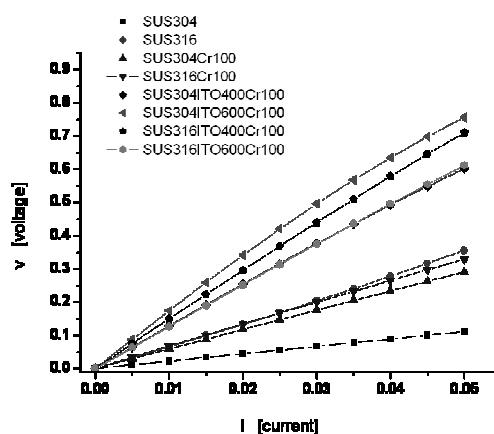


Figure 5: I-V characteristics of chromium film [100nm] and ITO film [400-600nm] coated-SUS 304, 316 plates.

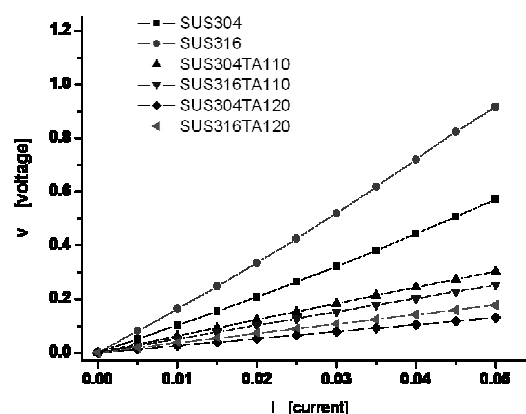


Figure 6: I-V characteristics of gold film [1-2um] and titanium film [0.1um] coated-SUS 304, 316 plates.

Fig. 6 shows the current-voltage characteristics of the stainless steel plates

deposited with the multi-layered coatings of gold-titanium system 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, were shown in Fig. 6. The thick gold film of 2 μ m thickness decreased the electrical resistance of the stainless steel plates modified with the multi-layered coatings. The stainless steel 304 plate modified with 2 μ m Au film and 0.1 μ m Ti film showed lowest electrical resistance in the current-voltage relationship (see Table 1). All the stainless steel plates deposited with gold-titanium system showed mostly low resistance values.

Table 1. Electrical resistance of stainless steel plates deposited with gold-titanium film and CrN-(Cr₂N)-chromium-ITO film.

[Unit : Ohm]

Specimen	SUS 304 bipolar plate	SUS 316 bipolar plate
Surface structure		
Non-coating	14.662	14.79
CrN+Cr ₂ N/ 100 nm Chromium film	5.804	6.562
CrN+Cr ₂ N/ 100 nm Chromium film / 200nm ITO film	14.232	11.9
CrN+Cr ₂ N/ 100 nm Chromium film / 400nm ITO film	12.046	14.19
CrN+Cr ₂ N/ 100 nm Chromium film / 600nm ITO film	10.08	12.222
1 μ m Gold film 0.1 μ m Titanium film	6.07	5.07
2 μ m Gold film 0.1 μ m Titanium film	2.61	3.54

3.4 Water contact angles of the metal bipolar plates modified with multi-layered coatings

Fig. 7 shows the contact angles of the bipolar plates coated with chromium film and intermediate ITO film. The stainless steel bipolar plates modified with CrN (Cr₂N)-chromium-ITO system showed a contact angle of about 80 and 90 degree. Fig. 8 shows the contact angles of the bare stainless steel plates and the bipolar plates coated with the multi-layered coatings of

external gold film and titanium film. The bare-SUS 304, 316 plates showed approximately 60-70 degree and the stainless steel bipolar plates modified with multi-layered coatings of gold-titanium system 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 μ m) and titanium film represented prominent hydrophobic behavior with average contact angles of 98.5 degree and 99 degree, respectively.

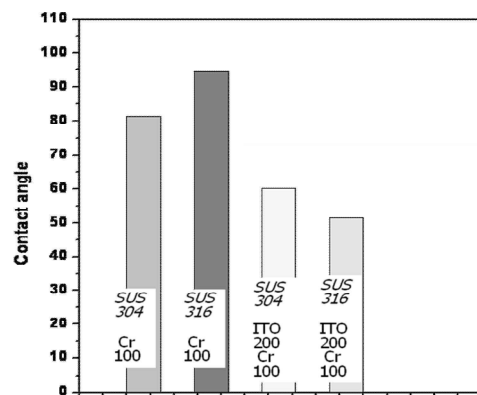


Figure 7: Contact angles of chromium film [100nm] ITO film [200nm] coated-SUS 304, 316 plates.

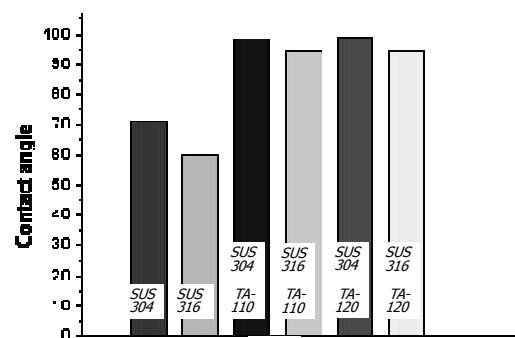


Figure 8: Contact angles of stainless steel 304, 316 plates deposited with Ti and Au. TA-110 (1 μ m Au / 0.1 μ m Ti), TA-120 (2 μ m Au / 0.1 μ m Ti)

High water contact angle of the bipolar plates in PEM fuel cell is required for the effective drainage or circulation of water created during cell operation. The multi-

layered coatings of gold and titanium as well as chromium increased remarkably the water contact angles of stainless steel plates

4. Conclusions

Two multi-layered coatings of the CrN (Cr₂N)-chromium-ITO system and the gold-titanium system have been deposited on stainless steel 304 and 316 plates, respectively. The XRD patterns of the plates modified with chromium-ITO system showed typical crystalline phases of the chromium metallic film, chromium nitrides (CrN and Cr₂N), ITO film and SUS substrate. The metallic bipolar plates coated with the gold-titanium system showed γ -Fe phase pattern of stainless steel as well as strong peaks of the Au phase (the external film) in XRD patterns. The multi-layered coatings of the CrN (Cr₂N)-chromium-ITO system showed the nano-scale columnar grains of the surface region in FE-SEM images and a grain size variation according to the thickness of the intermediate films. The surface morphology of the gold-titanium film formed on surface of stainless steel 304, 316 plates represented grains of about 100nm diameter. The multi-film deposited stainless steel bipolar plates modified with chromium-ITO film showed resistance of approximately 5-6 Ohm. The electrical resistance of the stainless steel plates, modified with gold-titanium film, decreased with the increase of the external film thickness from 1 μ m to 2 μ m. The stainless steel bipolar plates modified with gold-titanium film and chromium metallic film revealed high water contact angles in the range of 90 degree to 99 degree (average values) compared to that of the bare stainless steel plates.

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