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Characterization of multi-film deposited bipolar plates for a PEM (Proton Exchange Membrane) fuel cell application

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Two different films, an external metallic film (100 nm) and a conductive interlayer (200-600 nm) have been applied to the surface of stainless steel 316, 304 plates by a sputtering method and an E-beam method, respectively. The metal nitride film was formed on the external surface regions of the two-film deposited stainless steel plates through a simple annealing procedure. The XRD patterns of the stainless steel plates deposited with the metallic film and a conductive oxide film showed the crystalline phase of a typical indium-tin oxide, a metallic phase and the metal substrate as well as external nitride film. The nitride film was composed of two metal nitride phases of CrN and Cr_2N compounds. Surface microstructural morphologies of the multi-film deposited, stainless steel bipolar plates were observed by AFM and FE-SEM images. The metallic top film on the stainless steel plates showed a microstructural pattern composed of fine columnar grains with 10nm diameter and 60nm length in FE-SEM images. The electrical resistivity and contact angle of the coated-stainless steel bipolar plates were measured with the thickness of the oxide film.

Key words : Proton exchange membrane fuel cell (PEMFC), Bipolar plates, Conductive film, Metallic film, Metal nitride, Resistivity.

Introduction

The concept of using hydrogen as a new energy source has been suggested as a long-term solution to the problems of planetary warming and pollution, energy [1]. Great progress and persistent studies have been made since the early 1970s with activity concerning the hydrogen energy and hydrogen economy. Hydrogen economy has been developed very actively in all aspects: the production of hydrogen, storage and transportation, utilization in industry, in transport vehicles, and in everyday life. Recently, 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. Fuel cell technologies for vehicle applications are concerned with proton exchange membrane (PEM) fuel cells [4-7].

The present PEM fuel cells utilize the machined, very thick and heavy graphite blocks (bipolar plate). For commercialization of PEMFCs, it is required to reduce the manufacturing cost and weight of the fuel cell components [4, 8]. As alternative bipolar plate materials, carbon composites and metal plates have been studied for the PEMFCs [5, 7, 9]. 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 PEMFCs [9-13].

In this study, stainless steel 304 and 316 plate were selected as the metallic bipolar plate materials and coated with conductive and metallic films. The coating process for the oxide film and metallic film on bipolar plates was carried out by a sputtering method and an E-beam method. The crystal phases of the plate coated and bare stainless steels were identified from XRD patterns. The electrical resistivity of the stainless steel and the plates coated with conductive film and metallic film were measured. Surface morphologies of the bare stainless steel and the steel plates coated were observed by FE-SEM, AFM. Contact angles of the coated-stainless steel bipolar plates and the bare stainless steel plates was measured.

Experimental procedure

Commercial stainless steel 304 and 316 plates were selected as the substrates for bipolar plates for proton exchange membrane fuel cell (PEMFC) application. The surface of the stainless steel plates was cleaned with isopropylalcohol. The surface of the stainless steel 304, 316 plates was coated with the conductive oxide, indium-tin oxide film and chromium metallic thin film. The electrical resistivity of stainless steel plates was

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evaluated before and after the conductive coating procedure. The multi-film formation of the conductive oxide film and metallic film on stainless steel plates was conducted by a sputtering method (Vacuum Science Co. Korea) and an E-beam method (Electron Beam Evaporator, World Science Co., Korea). The crystalline phases of the multi-films 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 film and metallic film and the bare stainless steel plates were measured by an i-v source meter (Keithley, USA). The surface microstructural morphology of the multi-film coated stainless steel plates was observed by 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 multi-film deposited stainless steel plates were measured by a contact angle meter (Digidrop, GBX, France).

Results and Discussion

XRD patterns of the multi-film deposited steel bipolar plates

Figure 1 shows the XRD patterns of the metallic film and conductive interlayer deposited stainless steel bipolar plates and the as-received stainless steel plates. All the multi-film coated steel plates showed a typical γ-Fe phase pattern of stainless steel plates. The conductive film and metallic film coated-stainless steel plates showed the typical indium-tin oxide pattern and chromium pattern. Also, the multi-film coated bipolar plates indicated the presence of crystalline metal nitride

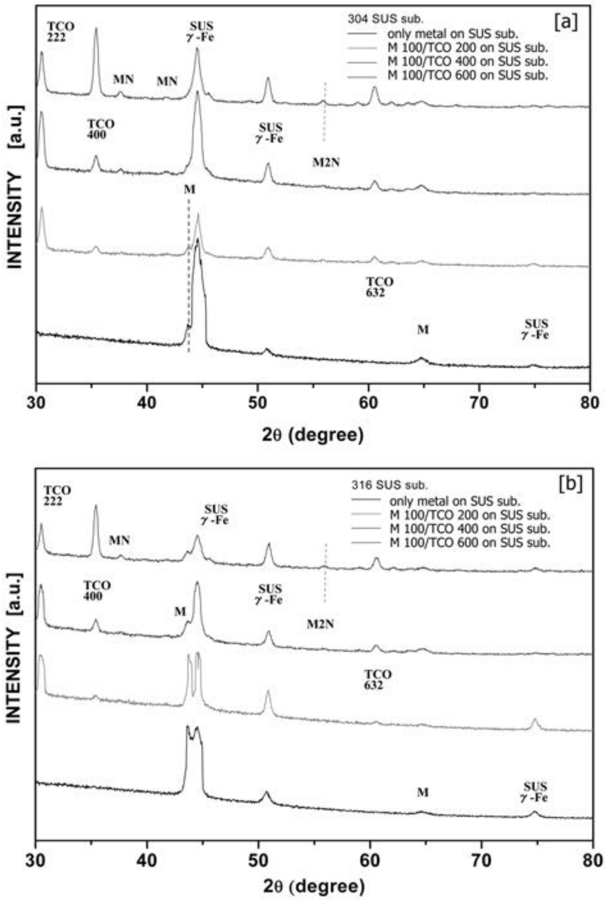

Fig. 1. (a) XRD patterns of metallic film [M: 100 nm] and conductive film [TCO: 200-600 nm] on stainless steel 304 plates. (b) XRD patterns of metallic film [M: 100 nm] and conductive film [TCO: 200-600 nm] on stainless steel 316 plates.

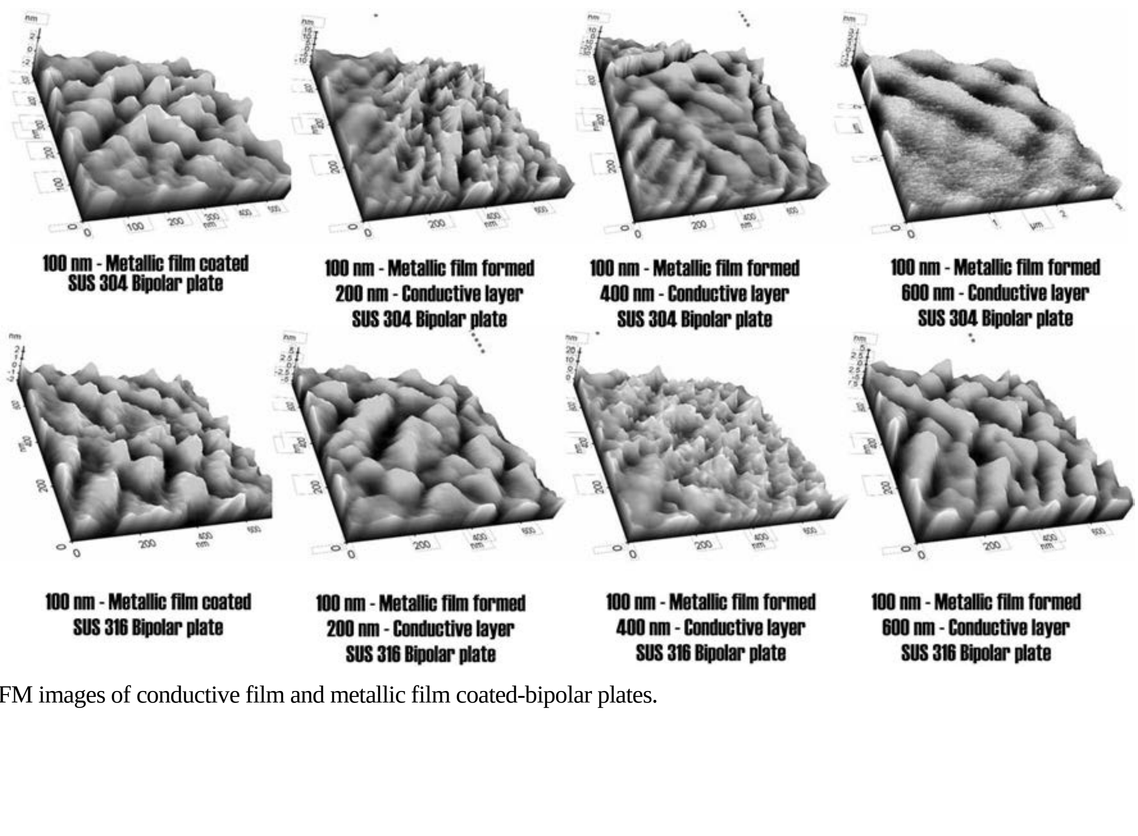

Fig. 2. AFM images of conductive film and metallic film coated-bipolar plates.

phases. It was inferred that chromium nitride compounds such as CrN or Cr_2N 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 resistivity 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. The multi-film coated stainless steel 304 plates indicated shrinkage of the metallic peak near 43 degree by increasing the conductive interlayer from 200 nm to 400 nm. The stainless steel 316 plates coated with a metallic film and conductive layer showed the metallic

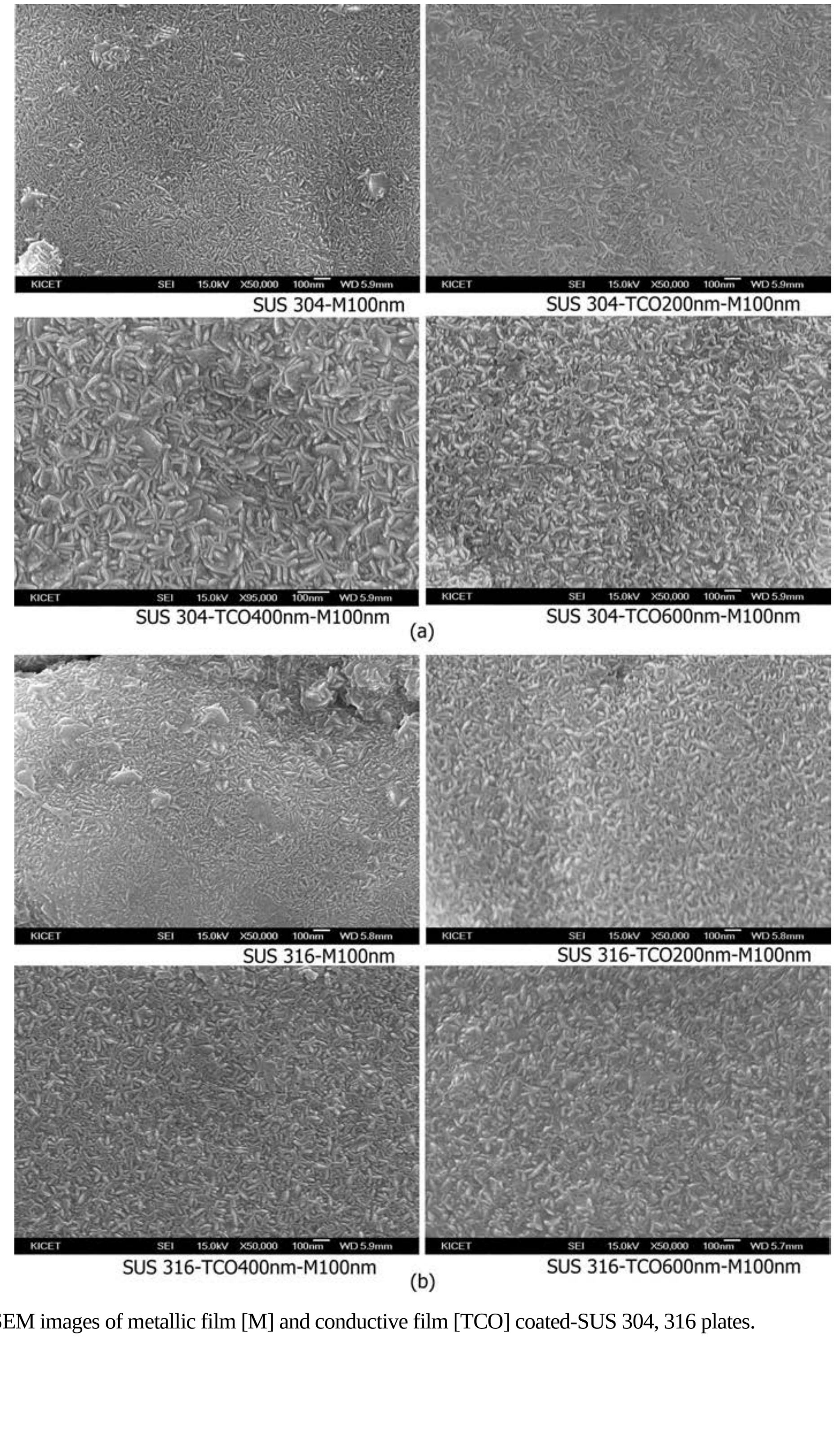

Fig. 3. (a), (b) FE-SEM images of metallic film [M] and conductive film [TCO] coated-SUS 304, 316 plates.

peaks near 43 degree for an interlayer thickness from 200 nm to 600 nm. The metal nitride phase such as CrN and Cr_2N compounds were shown mainly in the XRD patterns of the stainless steel plates coated with a metallic film and a conductive interlayer of 600 nm thickness.

Surface morphology of the multi-film deposited steel bipolar plates

Figure 2 shows AFM surface morphologies of the SUS bipolar plates coated with a topcoat of a chromium metallic film (100 nm) and an interlayer of conductive oxide film (200-600 nm). AFM images of the coated-steel plates showed a roughness variation with the oxide film thickness. The multi-film coated steel plates having a self-ordered dot pattern in the AFM image showed a very low roughness value of approximately 0.389 nm. Figure 3 shows FE-SEM images of the stainless steel bipolar plates coated with the top metallic film and conductive oxide film. 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 conductive film. The fine columnar grains of the external surface show a uniform shape of approximately 10 nm diameter and 60 nm 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. Although the multi-film deposited stainless steel plates and the steel plates coated with only the metallic film were composed of the metallic film and upper metal nitride film equally, then showed very different surface morphologies. In this study, it was found that the surface morphologies of the stainless steel bipolar plates coated were influenced by the presence of the interlayer.

Current-voltage characteristics of the multi-film deposited steel bipolar plates

Figure 4 shows the current-voltage characteristics of the bare-SUS 304, 316 plates and the multi-film coated-stainless steel bipolar plates. 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 bare-stainless steel 304 and 316 plates showed resistivities of approximately 2 ohmcm and 7 ohmcm, respectively. The multi-film coated SUS bipolar plates showed a resistivity of approximately 10 ohmcm. The stainless steel 304, 316 bipolar plates, which were modified with only the metallic film by the E-beam method, showed relatively low resistivities of about 5 ohmcm and 6 ohmcm, respectively.

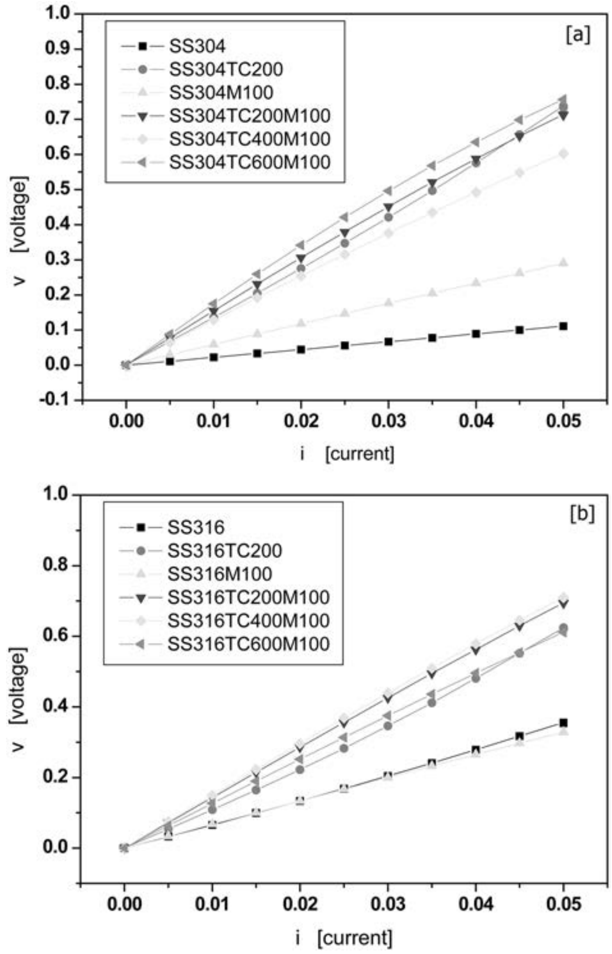

Fig. 4. (a), (b) I-V characteristics of metallic film [M: 100 nm] conductive film [TC: 200-600 nm] coated-SUS plates.

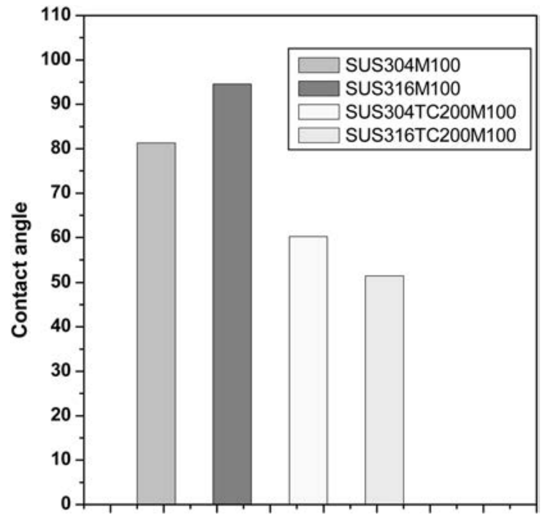

Fig. 5. Contact angles of metallic film [M: 100 nm] conductive film [TC: 200 nm] coated-SUS plates.

Contact angles of the multi-film deposited steel bipolar plates

Figure 5 shows the contact angles of the bare stainless steel plates and the bipolar plates coated with the metallic topcoat and the conductive intermediate film.

The bare-SUS 304, 316 plates showed approximately 60 degree and the stainless steel bipolar plates modified with two materials showed a contact angle of about 80 and 90 degree. The contact angle of the bipolar plates in PEM fuel cell is very important in the effective drainage or circulation of water created during cell operation. The bipolar plates for PEM fuel cell application require a high contact angle and a low surface energy.

Conclusions

A metallic film and a conductive oxide interlayer were formed on the stainless steel bipolar plates for PEMFC applications by E-beam method and sputtering method. The external film of metallic nitride phases was created by rapid thermal annealing. The thickness of the conductive oxide film and metallic film were approximately 200-600 nm and 100 nm, respectively. XRD patterns of the coated-SUS plates showed typical crystalline phases of the metallic film, metal nitrides, the conductive film and SUS substrate. Nano-scale columnar grains of the surface region were observed in FE-SEM images of the multi-film coated specimens and showed a size variation according to the deposition thickness of the intermediate films. It was evident that the presence of the conductive interlayer had an influence upon the surface morphologies and XRD patterns of the multi-film deposited stainless steel bipolar plates. The multi-film deposited stainless steel bipolar plates modified with a conductive film and a metallic film showed a resistivity of approximately 10 ohmcm and rather a high contact angle compared to that of the bare stainless steel plates.

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