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Durability and characterization studies of chromium carbide coated aluminum fuel cell stack

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ABSTRACT

Corrosion and interfacial contact resistance measurements were performed on chromium carbide coated aluminum 6061 and as-received aluminum 6061 samples. The coating was thermally sprayed onto the aluminum sample using the High Velocity Oxygen Fuel (HVOF) thermal spray technique. The chromium carbide coating consists of Cr₃C₂ top layer and Cr–C–Ni intermediate layer. The coating thickness was approximately 150 μm. A three-cell stack with chromium carbide coated aluminum bipolar plates was also fabricated for the durability and characterization studies. The coating thicknesses on the lands of the ribs and the walls of the valleys were approximately 300 μm and 150 μm, respectively. The stack was operated at the temperatures of 37 °C for 250 h and 80 °C for additional 500 h. The scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDX) analysis shows that the thickness, chemistry, and surface morphology of the coating material remained consistent after 750 h of operation. The inductively coupled plasma – optical emission spectroscopy (ICP-OES) analysis was performed on the samples of membrane electrode assembly (MEA) and byproduct water that was produced during the fuel cell electrochemical reaction.

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Introduction

Polymer electrolyte membrane (PEM) fuel cells convert chemical energy of hydrogen and oxygen gases into electrical energy. They are environmentally friendly and are ideal for residential and mobile applications due to their high power density, wide operating range, and low operating temperature [1–4]. Bipolar plates serve as the backbone of a PEM fuel cell stack. They isolate the individual cells, conduct current between cells, facilitate water and thermal management

through the cell, provide conduits for reactant gases, and facilitate the removal of reaction products [5,6]. Bipolar plates must be made of materials with high corrosion resistance, low interfacial contact resistance, high electrical conductivity, high mechanical strength, low permeability, no brittleness, and low cost. In addition, bipolar plates must have excellent manufacturability and suitable for cost-effective high volume production [7–11]. Wang et al. [12] projected 500,000 fuel cell stacks per year production in a Department of Energy annual progress report. This would result in millions of bipolar plates manufactured per year.

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Graphite is considered as one of the suitable materials for bipolar plate applications due to its excellent corrosion resistance, low interfacial contact resistance, chemical stability, and high thermal conductivity. However, brittleness, low mechanical strength, relatively high gas permeability, and high cost for machining gas channels limit its applications [13–16]. Although mechanical strength and brittleness of graphite can be improved by adding polymer fillers into the material, high polymer to carbon ratio can result in high interfacial contact resistance and low electrical conductivity. On the other hand, high carbon to polymer ratio will increase the composite's brittleness when compared to metallic plates [17]. This is not favorable in automobile industry where the bipolar plates are constantly under shock and vibration.

The main research goal for carbon composite plates is to improve electrical conductivity and mechanical strength of the plates. Lim et al. [18] developed a new technique to improve the electrical conductivity of carbon composite plates by spraying conductive particles such as natural graphite powder and carbon black onto composite prepreps before stacking. Their bipolar plates embedded with 10 wt.% of the carbon black had a flexural strength that was 9.4 times greater than the DOE target of 25 MPa. The bulk resistance of their plates was $4.95 \text{ m}\Omega \text{ cm}^2$ which was 50% less than that of conventional composite bipolar plates. Kang et al. [19] fabricated and tested composite bipolar plates with different graphite shapes (flake-like and lump-like), grades (natural and synthetic), and particle sizes (10, 20, 40 μm), and various resin contents (10, 15, 20, 25 wt.%). They concluded that flake-like natural graphite (NG) can be considered as the primary electrical conducting filler for composite bipolar plates due to the higher degree of crystallinity and aspect ratio of NG. Synthetic graphite (SG) can be used as the secondary conductive filler in order to enhance the flexural strength of the composite plates because it has a higher surface energy than NG. Their results also show that a larger graphite filler size is advantageous for increasing the electrical conductivity. However it reduces the flexural strength.

Metals are another material choice for bipolar plates because they provide excellent mechanical strength, high electrical conductivity, better durability to shocks and vibration, and more flexibility in fabrication when compared to graphite [1,10,13]. Metallic bipolar plates can be classified into coated and non-coated metals. Tawfik et al. [20] and Antunes et al. [21] conducted comprehensive surveys on metallic bipolar plate materials that have been utilized by many researchers. Stainless steel is one of the most studied metals in PEM fuel cell applications. One of the advantages of using stainless steel as a bipolar plate material is that the plate thickness can be very thin, which greatly reduces the weight of the fuel cell stack. For example, Dur et al. [22] studied the corrosion resistance behavior of 51 μm -thick SS316L bipolar plates. The plates were fabricated by stamping and hydroforming manufacturing methods. Their results indicated that coated-hydroformed bipolar plates exhibited higher corrosion resistance than the coated-stamped bipolar plates.

In general, bipolar plates made of bare stainless steel will experience passivation developing a passive film on the surface to prevent further corrosion. This passive film causes an undesirable effect of high interfacial contact resistance

resulting in the dissipation of some electrical energy into heat and a reduction in the overall efficiency of the fuel cell stack [23]. Various surface modification techniques have been investigated to minimize the passive film and yet maintain the high corrosion resistance of the stainless steel. For instance, Fu et al. [8] studied Cr-nitride films on SS316L. The corrosion test in a 0.5 M $\text{H}_2\text{SO}_4 + 5 \text{ ppm F}^-$ solution bubbled with air or hydrogen at 70 °C confirmed that the corrosion resistance of Cr-nitrided SS316L sample was greatly enhanced in comparison to the uncoated SS316L sample. The corrosion current densities of the Cr-nitrided stainless steel and uncoated stainless steel samples were about $10^{-6} \text{ A cm}^{-2}$ and $10^{-3.5} \text{ A cm}^{-2}$, respectively. The interfacial contact resistance of the Cr-nitrided SS316L sample was in the range of 8.4–11.8 $\text{m}\Omega \text{ cm}^2$ under the compacting pressure of 0.8–1.2 MPa, which was about two orders of magnitude lower than that of untreated SS316L.

Bai et al. [13] treated low carbon steel AISI 1020 by a reforming pack chromization process at a low temperature of 700 °C. They reported that a dense and homogenous Cr-rich layer was formed on the EDM activated carbon steels by pack chromization. The chromized coating on the low carbon steel had a low corrosion current density of $5.78 \times 10^{-8} \text{ A cm}^{-2}$ under 0.5 M H_2SO_4 solution and a low interfacial contact resistance (ICR) value of 11.8 $\text{m}\Omega \text{ cm}^2$ under 140 N cm^{-2} clamping pressure. Yang et al. [15] modified 316L stainless steel by the lower temperature chromizing treatment for bipolar plate application. Their results indicated that the chromized SS316L exhibited better corrosion resistance and lower ICR value than those of bare SS316L. The passive current density of the chromized SS316L was approximately $3 \times 10^{-7} \text{ A cm}^{-2}$ which was about four orders of magnitude lower than that of bare SS316L. The ICR value of the chromized SS316L was 13 $\text{m}\Omega \text{ cm}^2$ which was about one-third of bare SS316L's ICR value at 200 N cm^{-2} compaction pressure.

Aluminum is also a practical alternative for bipolar plate materials because of its low cost as compared to stainless steel and titanium. Also, an aluminum bipolar plate is much lighter than a stainless steel bipolar plate in weight if the bipolar plates are made of the same size. For instance, Lee et al. [2] reported that the weight of a 0.2 mm thick stainless steel bipolar plate is 83 g and the weight of an aluminum bipolar plate of the same size is just about 30 g, which indicates a 63.9% weight reduction. This magnitude of potential weight reduction has obvious advantages in transportation applications. To protect aluminum from the fuel cell corrosive environment, several surface treatments on aluminum bipolar plates have been reported for fuel cell applications.

Barranco et al. [24] conducted a corrosion test on a multi-layer chromium-zirconium nitride (ZrN–CrN) and a mono-layer chromium nitride (CrN) coated aluminum alloy plates (AL 5083) in a 0.5 M $\text{H}_2\text{SO}_4 + 2 \text{ ppm HF}$ solution at 70 °C bubbled with hydrogen or oxygen to simulate the anode and cathode environment respectively. The two coating materials were deposited on aluminum-based plates via cathodic arc evaporation physical vapor deposition (CAE-PVD). Their results showed that the corrosion current densities of the multilayer chromium-zirconium nitride coated aluminum plate (Al–CrN/ZrN) were $7.324 \times 10^{-5} \text{ A cm}^{-2}$ and $9.189 \times 10^{-5} \text{ A cm}^{-2}$ for the anodic and cathodic environments, respectively. The corrosion

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