

# Evaluation of coated metallic bipolar plates for polymer electrolyte membrane fuel cells

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## Abstract

Metallic bipolar plates for polymer electrolyte membrane (PEM) fuel cells typically require coatings for corrosion protection. Other requirements for the corrosion protective coatings include low electrical contact resistance, good mechanical robustness, low material and fabrication cost. The authors have evaluated a number of protective coatings deposited on stainless steel substrates by electroplating and physical vapor deposition (PVD) methods. The coatings are screened with an electrochemical polarization test for corrosion resistance; then the contact resistance test was performed on selected coatings. The coating investigated include Gold with various thicknesses (2 nm, 10 nm, and 1  $\mu\text{m}$ ), Titanium, Zirconium, Zirconium Nitride (ZrN), Zirconium Niobium (ZrNb), and Zirconium Nitride with a Gold top layer (ZrNAu). The substrates include three types of stainless steel: 304, 310, and 316. The results show that Zr-coated samples satisfy the DOE target for corrosion resistance at both anode and cathode sides in typical PEM fuel cell environments in the short-term, but they do not meet the DOE contact resistance goal. Very thin gold coating (2 nm) can significantly decrease the electrical contact resistance, however a relatively thick gold coating (>10 nm) with our deposition method is necessary for adequate corrosion resistance, particularly for the cathode side of the bipolar plate.

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## 1. Introduction

The bipolar plates for the polymer electrolyte membrane fuel cell (PEMFC) perform multiple functions, such as conducting electrons between adjacent cells, separating and distributing fuel and oxidant, and carrying away reaction products and heat from each cell. In order to perform these functions well, bipolar plates need to have low contact resistance and high electrical conductivity, high thermal conductivity, low gas permeability, good mechanical strength, satisfactory corrosion resistance, or suitable surface properties for liquid water removal. Traditionally, graphite and graphite composite plates with machined flow fields have been widely used in PEMFC because of their high corrosion

resistance, and low interfacial contact resistance (ICR). However, their poor manufacturability results in high fabrication cost, and their brittleness requires the use of fairly thick plates for adequate mechanical strength. Recently, metallic bipolar plates have received much attention as an alternative bipolar plate material because of their superior mechanical strength, low gas impermeability, very high bulk electrical and thermal conductivity, relatively low material and manufacturing cost [1]. Typically, bipolar plates are exposed to a warm (65–90 °C), acidic (pH 2–3) and humid environment. Under such conditions, the thermodynamically stable state of most of the metallic elements is frequently the cations, rather than the metal itself. For example, in the case of stainless steel bipolar plates, metal cations, such as  $\text{Fe}^{2+}$ ,  $\text{Ni}^{2+}$  and  $\text{Cr}^{3+}$  can be released during the corrosion process [2]. Additionally, poorly conductive metal oxides can form on the corroded surfaces, increasing the ICR of the contacting surfaces and thus reducing fuel cell performance.

The corrosion process is an electrochemical process which involves both oxidation and reduction reactions. In case of metal

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corrosion in acid solution, metal oxidation takes place at the anode sites, and reduction of hydrogen ions occurs at cathode sites [3–5]. The processes can be represented as follows:



In Eq. (1), M represents surface metal element,  $M^{n+}$  is the corresponding metal ion in the solution,  $e^{-}$  is free electron; in Eq. (2),  $H^{+}$  and  $H_2$  represent proton adsorbed on metal surface and the evolved hydrogen gas. Besides these, oxidation and reduction reactions involving other ions (ferrous and hydroxide ions) and species (oxygen) can also occur depending on the solution used.

The corrosion reactions involve the transfer of electrons and ions between the metal and the solution. Without an external applied potential or external source of electrons, surface corrosion reactions normally occur at a potential for which the rate of oxidation and reduction are electrochemically balanced. The equilibrium potential (corrosion potential,  $E_{\text{CORR}}$ ) and the associated current (corrosion current,  $I_{\text{CORR}}$ ) can be measured by an electrochemical polarization test.  $E_{\text{CORR}}$  and  $I_{\text{CORR}}$  are widely used as the indicators of the corrosion resistance of a material. A typical polarization curve for bare stainless steel (316) is shown in Fig. 1. The corrosion current and corrosion potential are determined by extrapolating the tangential line of the anodic and cathodic polarization curve.

Comprehensive research has been conducted on metallic bipolar plates and protective coatings over the past few years. An Fe-based alloy (FeB) and Ni-based alloy (NiB) were investigated for the suitability of cost-effective bipolar plates [6]. The polarization characteristics of the fuel cells with FeB plates tested indicated that most of the FeBs exhibited a characteristic behavior comparable to that of the Ni-based alloy (NiB) and was found to be susceptible to pitting corrosion. Moreover, contact resistance was about  $100 \text{ m}\Omega \text{ cm}^2$  at  $140 \text{ N cm}^{-2}$ , which was higher than that of NiB and does not meet the DOE targets (Table 1 [7]). Fleury et al. [8] evaluated an Fe-based amorphous alloy for the bipolar plates; the contact resistance of the Fe-based amorphous alloys were found to be similar to that of stainless steel and their

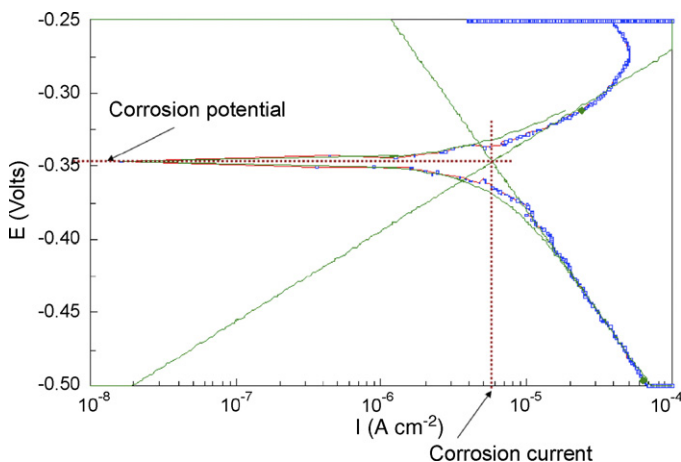


Fig. 1. Polarization curve of bare SS 316 around the corrosion potential in pH 2  $H_2SO_4$  solution at  $80^\circ C$  (Potential vs. SCE).

Table 1  
DOE targets for 2010; ICR, corrosion resistance, and cost

Performance metrics	DOE goal by 2010
ICR at $140 \text{ N cm}^{-2}$ ( $\text{m}\Omega \text{ cm}^2$ )	10
Corrosion current at $-0.1 \text{ V (H}_2)$ ( $\mu\text{A cm}^{-2}$ )	<1
Corrosion current at $0.6 \text{ V (air)}$ ( $\mu\text{A cm}^{-2}$ )	<1
Cost ( $\text{\$ kW}^{-1}$ )	6

corrosion resistance was poor in comparison with the graphite plates.

Stainless steel (SS) is the most widely used candidate for metallic bipolar plates and numerous reports have been published on this material by several institutes and universities over recent years. Stainless steel's relatively high resistance to corrosion, relatively low cost, and superior manufacturability make it an ideal base material for a host of commercial applications. Makkus et al. [9] conducted fuel cell testing with different stainless steel bipolar plates. The results show a current decay of approximately 30–50% in the first 300 h of operation, and a high metal ion concentration in the MEA. Similar work was performed by Davies et al. [10] with 310, 316 and 904L stainless steel bipolar plates. Li et al. [11] examined the corrosion behavior of 316 SS in a simulated anode environment. They stated that a coating needs to be developed to enhance the corrosion resistance of the stainless steel.

Some researchers investigated gold-coated bipolar plates, since gold, itself, is the one of the best corrosion resistance materials and an excellent electrical conductor. Hentall et al. [12] and Woodman et al. [13] fabricated current collectors from aluminum, then coated them with gold by a solution process. It was found that the fuel cell data with gold-coated bipolar plates showed a comparable performance to that of cells with graphite plates.

Wang et al. [14] conducted short-term fuel cell performance testing with gold-plated titanium bipolar plates. Since a long-term corrosion test was not implemented, feasibility of the bipolar plate was not completely validated. Surface contact resistance below  $1 \text{ m}\Omega$  was measured at a compaction torque in the range from  $20 \text{ kgf cm}$  to  $60 \text{ kgf cm}$ . Cell polarization curves indicated that the cell performance with coated plate was quite similar to that of graphite plates. Their cell was tested at  $40^\circ C$  with 5 psi back pressure; the humidification temperature of anode and cathode,  $90^\circ C$  and  $80^\circ C$ , respectively.

Most coatings for metallic bipolar plates have been proven inadequate due to pin-hole defects, which cause local corrosion and metallic ion contamination of the membrane. Recently, nitridation has been identified as an alternative coating method for metallic bipolar plates due to the combination of high electrical conductivity and good corrosion resistance [15]. The nitride layer is formed by nitrogen atom penetration and diffusion into the substrate, for which there is less interfacial adhesion problem, and is characterized by high corrosion resistance [16].

Brady et al. [15] have recently developed a preferential thermal nitridation process to generate a pinhole-free coating on a Ni–Cr alloy base plate, which shows excellent corrosion resis-

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