



Contents lists available at ScienceDirect

Journal of Energy Chemistry

journal homepage: www.elsevier.com/locate/jechem

Ti/(Ti,Cr)N/CrN multilayer coated 316L stainless steel by arc ion plating as bipolar plates for proton exchange membrane fuel cells

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ARTICLE INFO

Article history:

Received 8 June 2016

Revised 19 August 2016

Accepted 23 August 2016

Available online xxx

Keywords:

Stainless steel bipolar plate

Proton exchange membrane fuel cell

Arc ion plating

Multilayer coating

ABSTRACT

Arc ion plating (AIP) is applied to form Ti/(Ti,Cr)N/CrN multilayer coating on the surface of 316L stainless steel (SS316L) as bipolar plates for proton exchange membrane fuel cells (PEMFCs). The characterizations of the coating are analyzed by scanning electron microscopy (SEM) and X-ray diffraction (XRD). Interfacial contact resistance (ICR) between the coated sample and carbon paper is $4.9 \text{ m}\Omega \text{ cm}^2$ under 150 N/cm^2 , which is much lower than that of the SS316L substrate. Potentiodynamic and potentiostatic tests are performed in the simulated PEMFC working conditions to investigate the corrosion behaviors of the coated sample. Superior anticorrosion performance is observed for the coated sample, whose corrosion current density is $0.12 \mu\text{A/cm}^2$. Surface morphology results after corrosion tests indicate that the substrate is well protected by the multilayer coating. Performances of the single cell with the multilayer coated SS316L bipolar plate are improved significantly compared with that of the cell with the uncoated SS316L bipolar plate, presenting a great potential for PEMFC application.

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1. Introduction

Proton exchange membrane fuel cell (PEMFC) has attracted much attention over the past decades due to its high efficiency and zero emission [1]. It is promising for automotive, stationary and portable applications. Bipolar plate is the most bulky component in a PEMFC stack with respect to volume and weight [2,3]. Graphite is employed to fabricate bipolar plate currently for its excellent electrical conductivity and chemical stability. Graphite bipolar plate, however, is usually thick and heavy to ensure its low hydrogen permeability and high mechanical strength. What is more, the high cost of graphite bipolar plate hinders its commercial application in portable devices. Metal, especially stainless steel, is considered to be a promising candidate for bipolar plate fabrication due to its good bulk electrical conductivity, excellent mechanical strength, less gas permeability and cost effectiveness [4–6]. Unfortunately, the corrosion resistance of metal in the PEMFC working environments is inadequate. The releasing ions will future contaminate the membrane and poison the catalyst, besides, the interfacial contact resistance (ICR) between metal material and carbon

paper is high, which lowers the efficiency of PEMFC. Recent studies have revealed that bare metals could not be used to fabricate bipolar plate directly in PEMFC [7]. While a promising approach is to form a protecting coating with good corrosion resistance and low interfacial contact resistance on the surface of metal substrate. Noble metals are the most suitable candidates [8,9]. Nevertheless the high cost of the materials prevents them from commercial application. Metal nitrides are potential materials for surface modification such as Cr-nitrides owing to their good corrosion resistance, excellent interfacial conductivity and low cost [7].

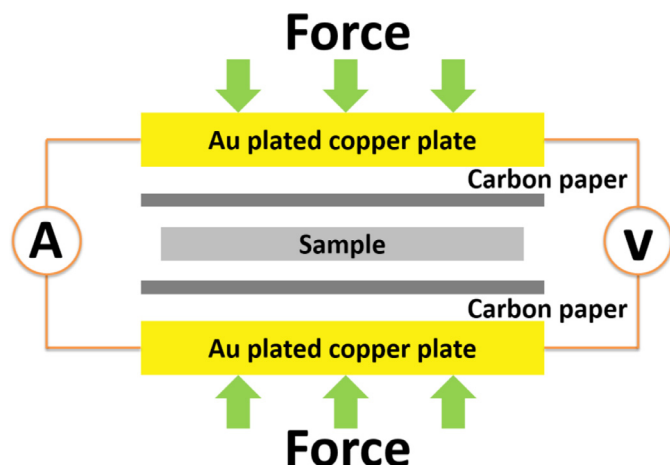
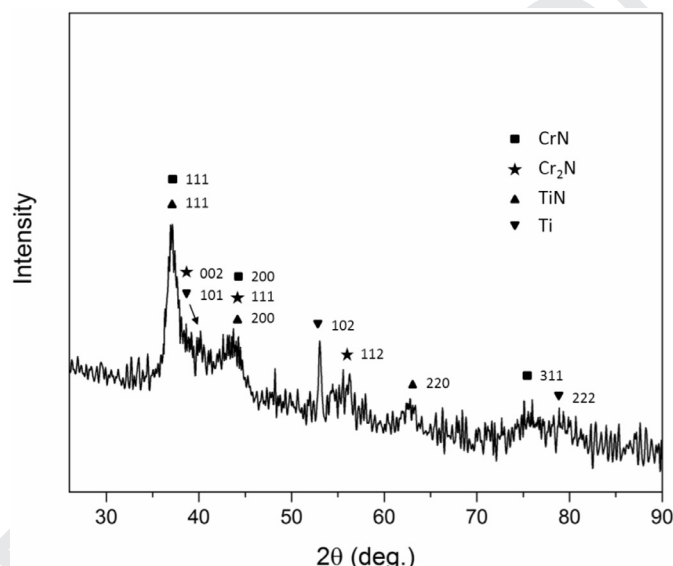
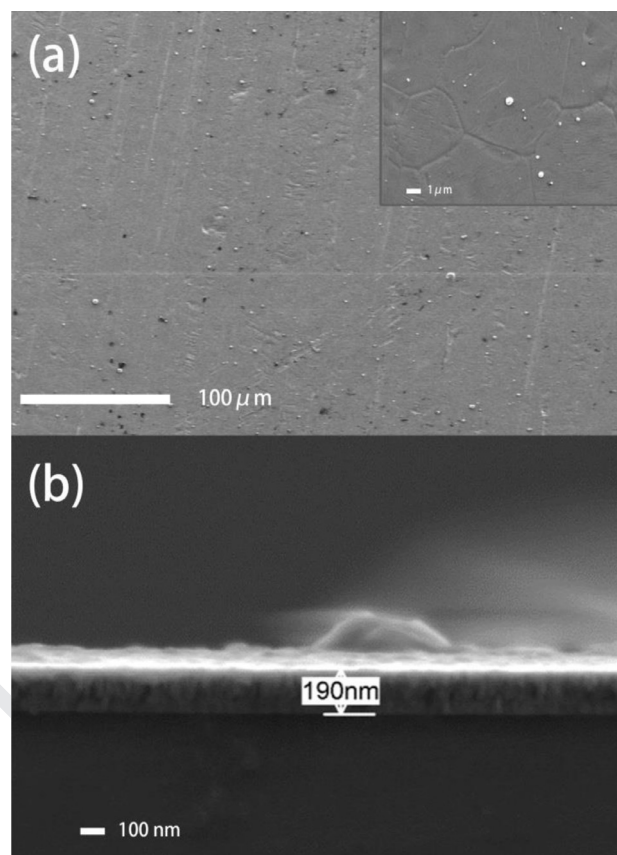
Hong et al. [10] formed CrN and Cr₂N compounds on the surface of SS316L by inductively coupled plasma, using a mixture of N₂ and H₂ at temperatures between 530 K and 650 K. Interfacial conductivity was improved significantly. While the corrosion resistance changed little owing to the formation of Cr-depleted regions. Brady et al. [11–15] Paulauskas et al. [16], Toops et al. [17] and Wang et al. [18–20] conducted a series of experiments to form Cr-nitrides coatings on different kinds of substrates including Ni–50Cr, AISI446, 349TM, Ni–Cr based, Fe–Cr based and Fe–Cr–V based alloys by thermal nitridation. ICRs were reduced for the nitrided samples. In addition, some specimens, especially the nitrided Ni–50Cr, the price of which is high, showed excellent corrosion resistance in both simulated anode and cathode working conditions. Similar works were done by Tian [21] and Lee [22,23].

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Table 1. Chemical composition of the 316 L stainless steel.

Elements	Fe	Cr	Ni	Mo	Mn	Si	P	C	N	S
Content (wt%)	Balance	16.67	10.15	2.12	1.348	0.576	0.0316	0.0212	0.0136	0.0021

**Fig. 1.** Schematic illustration of the interfacial contact resistance measurement equipment.**Fig. 3.** XRD pattern of the multilayer coating.**Fig. 2.** Surface morphology and profile SEM image of the multilayer coated SS316L sample.

[24] deposited CrN/Cr coating on the surface of SS316L by PVD technology. The coated sample exhibited improved corrosion resistance and surface conductivity compared with SS316L substrate under both simulated anode and cathode PEMFC operating environments. Lavigne et al. [25] assembled a stack with five single cells using CrN coated SS316L bipolar plates, which showed good performances after 200 h dynamic cycling. Ho et al. [26] investigated multilayer TiN/CrN coating deposited on SS316L substrate by cathodic arc deposition technique. They found that the multilayer coating deposited at rotation speed of 2 rpm showed the best corrosion resistance due to the multilayer structure and higher thickness of the coating. Barranco et al. [27] studied the corrosion behavior of CrN coated aluminum. Though the coated samples, the coating thicknesses of which ranged from 3 μm to 5 μm, showed superior performances than those of the as-received Al substrate in all cases, pitting holes were found in the coatings.

According to our previous studies [28–32], Cr-nitrides coating, prepared by arc ion plating technique with a device to reduce the droplets, exhibited good corrosion resistance and interfacial conductivity. In the present research, a layer of Ti, which is well known to have very high resistance to general corrosion and local corrosion [33,34], was deposited on the surface of the SS316L substrate to prevent corrosion from penetrating to the substrate through the flaws in the coating. Then a transition layer of (Ti,Cr)N was deposited before the CrN layer to improve the adhesion between the Ti layer and the CrN layer for the similar values of lattice parameters can improve the compatibility of the adjacent layers [35]. At last, a layer of CrN was deposited. The behaviors of the single cell with the multilayer coated SS316L bipolar plate were also studied. While the bare SS316L substrate was tested as reference. In addition, arc ion plating technique is cost effective to obtain dense films and realize low-temperature deposition.

2. Experimental

The austenitic 316L stainless steel was chosen as the base metal, the chemical composition of which is shown in Table 1.

Nevertheless, forming a continuous and external Cr-nitrides layer on the surface of the substrates, whose composition is strictly restricted, is arduous by thermal nitridation or plasma nitridation.

Physical vapor deposition (PVD) technology is a capable process to obtain the specific Cr-nitrides layer with less confinement. Tian

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