



# Electrochemical behaviour and surface conductivity of niobium carbide-modified austenitic stainless steel bipolar plate



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## HIGHLIGHTS

- Niobium carbide diffusion layer with dense structure is firstly prepared on 304 SS.
- The niobium carbide diffusion modified 304 SS bipolar plates are evaluated.
- Electrochemical results indicate that the niobium carbide diffusion layer has excellent corrosion resistance.
- The niobium carbide diffusion layer exhibits high surface conductivity even after corrosion tests.

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## ABSTRACT

A niobium carbide diffusion layer with a cubic NbC phase surface layer ( $\sim 6 \mu\text{m}$ ) and a Nb and C diffusion subsurface layer ( $\sim 1 \mu\text{m}$ ) is fabricated on the surface of AISI 304 stainless steel (304 SS) bipolar plate in a proton exchange membrane fuel cell (PEMFC) using plasma surface diffusion alloying. The electrochemical behaviour of the niobium carbide diffusion-modified 304 SS (Nb–C 304 SS) is investigated in simulated PEMFC environments (0.5 M  $\text{H}_2\text{SO}_4$  and 2 ppm HF solution at 80 °C). Potentiodynamic, potentiostatic polarisation and electrochemical impedance spectroscopy measurements reveal that the niobium carbide diffusion layer considerably improves the corrosion resistance of 304 SS compared with untreated samples. The corrosion current density of Nb–C 304 SS is maintained at  $0.058 \mu\text{A cm}^{-2}$  and  $0.051 \mu\text{A cm}^{-2}$  under simulated anodic and cathodic conditions, respectively. The interfacial contact resistance of Nb–C 304 SS is  $8.47 \text{ m}\Omega \text{ cm}^2$  at a compaction force of  $140 \text{ N cm}^{-2}$ , which is significantly lower than that of the untreated sample ( $100.98 \text{ m}\Omega \text{ cm}^2$ ). Moreover, only a minor increase in the ICR of Nb–C 304 SS occurs after 10 h potentiostatic tests in both cathodic and anodic environments.

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## 1. Introduction

High performance fuel cells have received increasing attention as alternative power sources due to the adverse environmental impact and decreasing reserves of fossil fuels. Unlike internal combustion engines, in which electric energy is produced from thermal and mechanical energy, fuel cells generate electric energy through electrochemical reactions using a combination of fuel and oxidants. As the most important type of fuel cell, the hydrogen fuelled proton exchange membrane fuel cell (PEMFC) is undergoing rapid development as a power source for mobile applications, especially for electric vehicles, due to its high efficiency and power density, low operating temperature and environmentally friendly properties [1–4]. As a multifunctional component of PEMFCs,

bipolar plates play an important role in connecting the anode of one cell to the cathode of the adjacent cell, supporting the cell stack, collecting current and providing a flow channel for fuel and oxidants. Previous reports have demonstrated that bipolar plates account for approximately 60–80% and 29–45% of the total weight and the total cost of the PEMFC stack, respectively [5–7]. Hence, it is necessary to explore light and low-cost bipolar plates to reduce the total cost and mass of PEMFCs and improve their appeal as commercial devices. Table 1 outlines the specific targets established by the U.S. Department of Energy (DOE) to be achieved by 2015 for bipolar plates in PEMFC stacks for automotive applications [8,9].

Stainless steel is widely accepted as one of the leading PEMFC bipolar plate material candidates due to its suitable physical and mechanical properties as well as relatively low cost [10–12]. However, the corrosion resistance of stainless steel is insufficient for successful application in a commercial PEMFC because the bipolar plates are subjected to acidic environments at elevated temperatures for extended periods of time. Some reports revealed

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**Table 1**  
U.S. DOE technical targets for bipolar plates set for the year 2015.

DOE technical targets: bipolar plate	
Cost (\$ kW <sup>-1</sup> )	3
System weight (kg kW <sup>-1</sup> )	<0.4
H <sub>2</sub> Permeation flux (cm <sup>3</sup> s <sup>-1</sup> cm <sup>-2</sup> )	<2 × 10 <sup>-6</sup>
Corrosion (μA cm <sup>-2</sup> )	<1
Electrical conductivity (S cm <sup>-1</sup> )	>100
Interfacial contact resistance at 140 N cm <sup>-2</sup> (mΩ cm <sup>2</sup> )	<10
Durability with cycling (h)	>5000
Flexural strength (MPa)	>25

that metal dissolution (in quantities as low as 5–10 ppm) from the metallic bipolar plates can contaminate the membrane electrode and diminish the performance of the PEMFC [13,14]. Moreover, the passive film formed on stainless steel surfaces in PEMFC environments would increase the interfacial contact resistance (ICR) between the bipolar plate and the gas diffusion layer (GDL), reducing the power output of the fuel cell stack. To address both the insufficient corrosion resistance and high surface resistivity of stainless steel, its surface can be modified with a protective coating. Ideally, the protective coating would be stable and impermeable to reactant gases and provide high conductivity as well as good corrosion resistance to withstand the aggressive PEMFC environment. Noble metals such as gold [15] and silver [16] are suitable coating materials due to their high corrosion resistance and high conductivity, but the commercial use of these materials, even in very thin films, is cost-prohibitive. Transition metals and their nitrides (e.g., Nb, Ta, ZrN, CrN, TiN, Ti/TiN, Ti/CrN, Cr/CrN/Cr) have been widely employed as protective layers for stainless steel bipolar plates using surface treatment techniques, including rolling cladding, magnetron sputtering, electroplating, chemical vapour deposition (CVD) and physical vapour deposition (PVD) [17–25]. Like nitrides, transition metal carbides also possess pronounced physical and chemical properties, such as superconductivity, high hardness, high melting points, high electrical conductivity and good corrosion resistance, making them ideal materials for use as wear resistance and corrosion resistance coatings, field emitters and diffusion barriers [26–31]. In particular, niobium carbide, as a refractory ceramic, exhibits excellent chemical stability and high conductivity. It was reported that niobium carbide exhibits a much higher electrical conductivity than ZrC and TiC [32–35]. Considering these advantageous properties, it is anticipated that niobium carbide may be a suitable protective coating for bipolar plates. However, no prior studies have explored the application of niobium carbide as a coating on bipolar plates. Therefore, in this study, a niobium carbide diffusion layer prepared by plasma surface diffusion alloying was applied as protective layer on an AISI 304 stainless steel (304 SS) bipolar plate. The electrochemical properties and surface conductivity of 304 SS with a niobium carbide diffusion layer (denoted as Nb–C 304 SS hereafter) were investigated in simulated PEMFC environments.

## 2. Experimental

Commercial austenitic AISI 304 SS sheets with thicknesses of 1.5 mm were employed as the substrate material. The composition of 304 SS is 0.049 C, 18.20 Cr, 8.66 Ni, 0.58 Si, 0.007 S and 0.021 P (wt.%), with the balance comprising Fe. The niobium carbide diffusion layer on the 304 SS sheet was prepared in a double glow plasma alloying furnace using a high purity Nb sinter plate as the sputtering source electrode, the 304 SS samples as the negative cathode and the furnace as the anode. The surface of the substrate was cleaned and heated using argon ion bombardment in a

vacuum chamber filled with pure Ar at a pressure of 40 Pa and a bias voltage of –1 kV. The source electrode power supply was loaded and maintained at –800 V while the bias voltage of the samples was decreased to –500 V. The samples were treated at a temperature of 1173 K for 1 h for the niobium diffusion process, and a proportional volume of acetone gas was then introduced into the chamber as a carbon source. Finally, the samples were treated for another 3 h to form the niobium carbide diffusion layer on the surface of 304 SS.

The crystalline structure of the Nb–C 304 SS was verified by X-ray diffraction (XRD). The surface and cross-sectional morphology as well as the thickness of the niobium carbide diffusion layer were analysed using a scanning electron microscope (SEM, JEOL JSM–6360LV) equipped with an Oxford energy-dispersive X-ray analysis spectrometer (EDS) system. The method reported by Wang et al. [36] was used to measure the interfacial contact resistance (ICR) at different compaction forces. The procedure is described in detail in Refs. [11] and [36].

The corrosion properties of Nb–C 304 SS and untreated 304 SS samples (with an area of 1 cm<sup>2</sup>) were characterised by electrochemical tests, i.e., potentiodynamic, potentiostatic polarisation and electrochemical impedance spectroscopy (EIS). The tests were conducted using a CHI660C electrochemical workstation controlled by a computer. To simulate the aggressive PEMFC operating conditions, the electrochemical tests were performed in a sulphuric acid aqueous solution (0.5 M H<sub>2</sub>SO<sub>4</sub> and 2 ppm HF solution at 80 °C) bubbled with either H<sub>2</sub> or air to simulate the anodic or cathodic environment, respectively. The three-electrode system comprising a saturated calomel electrode (SCE, as the reference electrode), a platinum sheet (as the counter electrode) and the samples (as the working electrode) was constructed for the electrochemical measurements. The EIS tests were carried out at open circuit potential in a frequency range from 100 kHz to 0.01 Hz with amplitude of 10 mV. To quantify the dissolved metallic Fe, Ni, Cr, and Nb ions produced in the potentiostatic process, the test solutions (approximately 100 ml for each test) were collected following 10 h potentiostatic tests, respectively. Inductively coupled plasma atomic emission spectrometry (ICP–AES) (Optima 2000 DV) was used to analyse the metallic ions dissolved in the collected solutions.

X-ray photoelectron spectroscopy (XPS) was employed to identify the superficial composition of the Nb–C 304 SS before and after potentiostatic polarisation. The XPS analysis was conducted in a K-Alpha 1063 (Thermo Fisher Scientific) electron spectrometer using an Al K $\alpha$  source (1486.8 eV) and a hemispherical energy analyser. To obtain information about the chemical composition throughout the depth of the niobium carbide diffusion layer, Nb–C 304 SS samples (before and after potentiostatic tests) were sputtered with argon ions. The sputtering rate was approximately 0.01 nm s<sup>-1</sup> for a period of 250 s, and X-ray photoelectron data were collected every 50 s, corresponding to a thickness of approximately 0.5 nm for each recorded layer. In addition, the thickness of the passive film, if present, can be calculated using the chemical state variation of the detected elements.

## 3. Results and discussion

### 3.1. Characterisation of the niobium carbide diffusion layer

The crystal structure of the niobium carbide diffusion layer was verified by X-ray diffraction (XRD). The XRD patterns of Nb–C 304 SS are displayed in Fig. 1. As observed in Fig. 1, only the cubic NbC phase (ICDD card no. 38-1364) with a dominant (111) preferred orientation is observed in the XRD pattern of Nb–C 304 SS, and no peaks from the 304 SS substrate were observed because the

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