



# An electrochemical treatment to improve corrosion and contact resistance of stainless steel bipolar plates used in polymer electrolyte fuel cells



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## HIGHLIGHTS

- Treatment for improving corrosion/contact resistance of stainless steel bipolar plates.
- Contact resistance of 316 stainless steel is reduced by over 800% at  $140 \text{ N cm}^{-2}$ .
- Treatment found to enhance Cr concentration in the near-surface.

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## ABSTRACT

An electrochemical surface treatment is presented that improves the properties of stainless steel (316SS) used as bipolar plates for polymer electrolyte fuel cells (PEFCs). The process is an anodic treatment, whereby the material is polarised beyond the transpassive region. Potentiodynamic corrosion testing, chemical and morphological surface characterisation and interfacial contact resistance measurements indicate that the improved properties of 316SS are primarily a consequence of an enrichment of Cr at the near-surface of the material. The surface treatment increases the corrosion resistance and significantly reduces interfacial contact resistance.

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## 1. Introduction

The polymer electrolyte fuel cell (PEFC) has shown great promise as a power source for stationary and transportation applications, largely due to its high power density and low temperature (typically 60–80 °C) operation. Although great strides have been made in the development of the various components for the PEFC, factors such as material performance, durability and production costs continue to retard development of fuel cells. The bipolar plate is an example of a component that still requires

significant improvement in order to optimise performance and realize operating targets [1].

The bipolar plate must carry fuel and oxidant to the respective electrodes and disperse the fluid in an even and controlled manner, it must conduct electronic current from each electrode, provide mechanical support and strength to the membrane electrode assembly (MEA), remove excess product water from the reaction zone, help control the temperature of the fuel cell by acting as a heat sink and physically separate reactant streams.

The cost of bipolar plate materials and processing required during manufacture make it one of the most expensive parts of the fuel cell. The bipolar plate typically accounts for 80–85% of the weight and most of the volume of fuel cell stacks [2]. Hence, low cost and lightweight materials are needed which do not compromise performance and durability.

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The multiple roles of the bipolar plate and the challenging environment in which it operates mean that the ideal material should be able to operate in an acidic environment (pH 3–5) in the presence of  $F^-$  ions (leached from the electrolyte membrane – particularly during start-up), potentials of 0–1.6 V vs. RHE (reversible hydrogen electrode) [3] and a temperature range of 60–120 °C [4]. The ideal bipolar plate should incorporate excellent corrosion properties, resistance to ion leaching, minimum electrical resistivity, be chemically and thermally stable as well as have good thermal conductivity, high mechanical strength and have a low contact resistance with the gas diffusion layer (GDL). Furthermore, they need to be thin and lightweight to increase the volumetric and gravimetric power density, be inexpensive and easy to fabricate as well as being environmentally benign in terms of manufacture and ultimate disposal/recycling.

Due to the need to maximise the power densities of PEFCs and lower the cost of construction, metallic bipolar plates are a popular choice for this component, albeit with the challenge of corrosion. Among many candidate materials based on metals and alloys, stainless steel has attracted much attention [5]. The advantages of using stainless steel are low cost, high strength and ease of incorporating a flow field via stamping or embossing. However, stainless steels are prone to chemical attack in the PEFC environment and corrosion products may poison the catalysts and/or ionomer membrane [6], which can have a significant effect on PEFC stack efficiency and cost [7].

In order to overcome the shortcomings of stainless steel with regards to corrosion, various surface coating and modification techniques have been proposed, including chemical vapour deposition (CVD) [8], physical vapour deposition (PVD) [9], electroplating [10] and nitriding [11]. It has also been reported that Cr-enriched surfaces can be beneficial to the interfacial contact resistance (ICR) [12].

The performance of stainless steel is limited by its surface characteristics, one of the issues being the voltage drop at the interface with the GDL due to the contact resistance associated with insulating surface oxides [1]. The nature of the surface oxide is a function of the alloying elements and the environment in which it operates inside the fuel cell. Studies by Davies et al. identified 316, 310 and 904L as promising grades of stainless steel based on the contact resistance and polarization characteristics [13]. It was noted that an increased Ni and Cr content in the alloy leads to lower interfacial resistance due to the formation of a thinner passive film. Wang et al. also investigated the applicability of different austenitic stainless steel compositions for bipolar plates by simulating the chemical environment of PEFCs [5]. Similarly it was concluded that decreasing contact resistance and corrosion current were observed for steels with increasing Cr content. The performance generally increases in the order 349TM > 904L > 317L > 316L. The results of these studies confirmed that Cr content plays a very important role in reducing ICR and consequently improves the performance of PEFCs.

Electrochemical processes resulting in chromium surface enrichment have been reported under traditional electropolishing conditions [14–16], as well as improving the passivity of iron-chromium alloys in dilute acid conditions [17]. Lee et al. developed an electrochemical process to increase the Cr activity at the surface of stainless steel to obtain a corrosion-resistant oxide [18]. Kim et al. demonstrated using specimens that underwent surface modification by immersion in acidic solution followed by heat treatment that rougher surfaces showed a lower ICR [19].

Electropolishing involves the dissolution of metal from the surface to produce a uniquely advantageous surface finish compared to the cutting, tearing and smearing action of abrasives and cutting tools which deforms the near-surface material, generating significant levels of residual stress. It may be used in lieu of

abrasive fine polishing in microstructure preparation [20]. Electropolishing is an effective means of passivating stainless steel, for example electropolished steel parts have been stored at 60–70% relative humidity for six months without visible rust [21]. It is a method for achieving new surface qualities that are outstanding in many applications while ensuring cost saving and technical improvements are realised. In addition to the decrease in the corrosion current density due to the formation of a passive surface film enriched with chromium, greater heat reflectivity and better emissivity may also be achieved [14].

Electropolishing is normally carried out in concentrated acid media and the polishing process is thought to involve the formation of a viscous layer at the metal surface, which can be enhanced by employing viscosity modifiers such as glycerol [14,20,21]. While electropolishing is a well-established industrial process there are issues associated with the technology, most notably that the solution used is highly corrosive and toxic, with extensive gas evolution occurring during the process.

Electropolishing is generally classified into two processes, surface levelling/roughening (etching) and surface brightening [22,23]. Electroetching is the process of surface roughening, with a matt finish result characterised by shallow pitting [15]. Etching is commonly performed on stainless steel (e.g. 0.05 A cm<sup>-2</sup> in 0.5 M Na<sub>2</sub>SO<sub>4</sub> under alkaline conditions) [24]. Brightening is commonly carried out in co-acidic electrolytes of H<sub>2</sub>SO<sub>4</sub> and H<sub>3</sub>PO<sub>4</sub>, with glycerol commonly used as an additive to suppress the influence of the metal microstructure on the dissolution rate [23].

Voltage transients under galvanic control can be used as an indicator of whether etching or brightening is taking place during anodisation treatments. At low current densities, when the dissolution voltage remains constant, the whole electrode is etched, while the formation of peaks at higher current densities usually corresponds to brightening [25].

The main objectives of the electropolishing process are: firstly, to remove the solid (Beilby) layer that consists of inclusions of martensitic phase, foreign material, and pre-existing oxides created by forming, machining or abrasive polishing [14,26]. Secondly, to create a new corrosion resistant layer that is enriched in chromium oxide due to anomalous co-dissolution and thirdly, to equipotentialize the grain boundaries of metallic materials [16].

While the electropolishing process is generally performed in quiescent electrolytes, flow dependent current plateaus that accompany electropolishing are observed [14,27]. The use of high current densities is also typically associated with the process. Although a low current is desirable in order to ensure a high current efficiency, consequently reducing hydrogen evolution and the flammability hazard [25], use of dilute electrolytes would help reduce this risk.

Sulphate-containing electrolytes are preferred for stainless steel dissolution as many reducing agents, which produce sulphate anions, are available to convert Cr<sup>6+</sup> to Cr<sup>3+</sup> [15]. Furthermore, the use of dilute sulphate electrolytes at low current densities has been shown to bring about improved surface qualities [25].

The rate determining process during electropolishing is diffusion of product ions away from the surface. The properties of this diffusion layer are affected by its viscosity, which can be used as a means of controlling the process. The viscosity of the polishing film may be modified by addition of a viscosity enhancing agent, typically a polyol, e.g. glycerol (propane-1,2,3-triol). On this basis, the preferential dissolution of ions from microscopic protrusions on the surface of the metal can be accounted for by the difference in the concentration gradient over the ‘peaks’, where the diffusion layer is thin and the concentration gradient is high, and ‘valleys’ where the diffusion layer is thicker and concentration gradient is lower.

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