IMPROVING THE CORROSION BEHAVIOUR AND DURABILITY OF METALLIC BIPOLAR PLATE FOR THE PEM FUEL CELL APPLICATION

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ABSTRACT

Proton Exchange Membrane Fuel Cell (PMFC) is known for its pollution free power generator in the transport application. Bipolar Plate or the current collector (BP) is a major part in the Proton Exchange Membrane Fuel Cell (PEMFC) and has the excellent electrical conductivity more than $>100\text{Scm}^{-1}$, flexural strength higher than 25Mpa and better corrosion resistance more than $1 \times 10^{-6} \text{Acm}^{-2}$ as per the Department of Energy (DOE, US). Earlier, graphite based bipolar plate are used and are replaced due to their high cost and poor machinability. Graphite bipolar plate is replaced by the metallic plate especially stainless steel, as they are most cost effective and easy to machine. Moreover, stainless steel bipolar plate was corroded in the acidic medium and its durability was detained. Recent research works were focused mainly on the selection of costless and effective coating techniques for the BP in PEMFC environment. This research paper reveals an inclusive review of various surface coatings and surface tailoring techniques for the stainless steel BP for enhancing the electrical conductivity and corrosion resistance property in PEMFC application.

KEYWORDS: Metallic Bipolar Plate, Corrosion Resistance, Stainless Steel, Surface Coatings & Polarisation Tests

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1. INTRODUCTION

Energy is claimed as the most powerful source in our day today industrial sectors. Almost majority of energy is obtained from fossil fuels. As far as the fossil fuels were concerned, petroleum is considered to be the most sought after fuel in the industrial economy. Few countries has plenty of petroleum resources to supply to other nations, where as many countries were lacking it and has failed to meet their demand. Finally, due to the demand for petroleum products in the future, the energy is obtained through renewable source like solar, wind, tidal, hydro and biomass. As the energy consumption rate is increasing more in the world economy, this renewable energy will not be a sufficient source. As far as the energy issue is concerned, the development of fuel cells was initiated at a higher rate.

Fuel cell comprises of anode and cathode at each side with an electrolytic solution as the medium. The inlet fuel gets oxidized at the anode portion, whereby the oxygen reduces on the cathode region with the by product having heat and water. The fuel cells like PAFC, AFC, PEMFC and DMFC operate at a lower temperature, whereas MCFC and SOFC works with higher temperature. General Electric Corp. in association with NASA has developed the Proton Exchange Membrane Fuel Cell (PEMFC) in space application. This fuel cell produces electricity through the chemical reaction between the fuel gas (H_2) at anode and oxygen (O_2) at the cathode with heat and water as the by-product.

Figure 1: Proton Exchange Membrane Fuel Cell.
(Source -en.wikipedia.org)

The PEMFC is made up of proton exchange membrane, platinum as the catalyst, gas diffusion membrane, bipolar plate, anode and cathode, as shown in the figure 1. The functions of bipolar plate inside the fuel cell is to disperse the reactants equally over the entire cell, dividing them in the cell stack, heat exhausting, current collecting and distributing them across the stack, through the superior electrical conductivity ($>100 \text{ Scm}^{-1}$), flexural strength ($>25 \text{ MPa}$) and better corrosion resistance ($<1 \times 10^{-6} \text{ Acm}^{-2}$) as per the Department of Energy (DOE, US) as presented below in table 1.

Table 1: Target Datas for the Bipolar Plate as per the DOE, US

Parameters	Target Values of DOE
Electrical Conductivity	$>100 \text{ Scm}^{-1}$
Tensile Strength	$>41 \text{ MPa}$
Flexural Strength	$>21 \text{ MPa}$
Corrosion Resistance	$<1 \times 10^{-6} \text{ A/cm}^{-2}$
Thermal Conductivity	$>10 \text{ W/m}^{-1} \text{K}^{-1}$
Interfacial Contact Resistance	$<20 \text{ m}\Omega \text{cm}^{-2}$ at 150Pa

Figure 2: Bipolar Plate Made of Stainless Steel.
(Source – h2fc-fair.com).

Graphite based BP usage was limited due to its poor machinability, low strength with high cost. Many researchers has preferred metallic BP than the carbon based, as the metals have higher mechanical strength, good machinability and cost effective. Hybrid Aluminium matrix composites (HAMC) is one of the classical material and can be used for the manufacturing of BP with superior engineering properties with low cost and easily machinable nature [31]. Titanium, copper alloys and nickel alloys are also made as the materials for metallic bipolar plate. From the survey, stainless steel is

considered as the better alternate material for bipolar plate application, as shown in the figure 2.

Bipolar plate inside the cell stack will lose its durability by the degradation of the plate due to corrosion medium and the lack of mechanical strength causes fracture. The performance of the fuel cell is dependent on the water management of the entire cell. When they are stagnated inside the stack, they react with the membrane and form an acid medium, and in turn affect the bipolar plate. Corrosion happens at both sides of the stack. On the cathode side, they enhance the growth of metal oxide layers, which may raise the electrical contact resistance and increases the electrical resistance of the plate. At the anode end, the reduction of metal has the ability to attack the polymer membrane and drags down the durability of the cell. Further, the catalyst is disturbed by the formation of the hydrides at this point. From the corrosion attack, these plates lose their mechanical strength and get fractured during the cell operation, as shown in the figure 3. Thermal stability of the bipolar plate will be disturbed due to the poor mechanical strength and may lead to erosion as the cell's temperature is around 100°C.

Figure 3: Damaged Stainless Steel Bipolar Plates under the effect of Corrosion.

(Source – en.wikipedia.org)

Recent trends were focused mainly on the selection of costless and effective coating techniques and also alternate materials for the BP for eradicating such corrosive effect in PEMFC environment. Low cost, easily available and high quality based surface coating technology is the current need. Surface engineering is an approach of tailoring any engineering material's texture and enhancing their properties and durability [32-39].

From the literature survey, there is lighter on the different coating process and surface tailoring techniques for stainless steel BP for the betterment of both the electrical conductivity and corrosion resistance. Many researchers focussed on different coatings like Physical Vapour Deposition (PVD), Chemical Vapour Deposition (CVD) electroplating, electroless plating, thermal spraying, pack cementation aluminizing, cladding, carburizing and nitriding. Among them, the metal nitride coating has more corrosion resistance than others. This paper provides a complete review of various anti corrosive coatings and surface modification method like electro plating, plasma nitriding, PFD, CVD, dip coating, pack cementation through different researchers on stainless steel BP for improving the electrical conductivity and corrosion resistance as per the target values from the DOE, US.

2. DESCRIPTION

The literature studies were focussed mainly on the various surface coating techniques that were deployed on the bipolar plate made from stainless steel in PEMFC. Lin-Chang Tsai et al. performed CrC and Ni electroplating followed by low temperature pack chromizing on the steel substrate. His result showed good improvement in corrosion rate in BPP for

PEMFC [1]. Yu Fu et al. has produced carbon coated 316L SS BP through the pulsed bias arc ion plating process. He has established three fine layers such as pure C, C-Cr composite layer and C-Cr-N layer. From the corrosion test, he has concluded that C-Cr film has showed the superior corrosion potential value than the bare sample [2]. The incorporation of Ti and C over the 316L SS BP through the ion implantation technique has enhanced the electrical conductivity and ICR is detained by 6.5 times. Kai Feng et al. have revealed that the corrosion resistance was gradually increased due to the combined effect of Ti and C ions [3]. Jun –Ho-Kim et al. has improved the ICR and corrosion property of the 316L SS BP through the inclusion of niobium ions over the SS surface by the sputtering method. The uniform dispersion of the Nb ion has enriched the ICR value than the bare substrate with appreciable corrosion potential [4]. Through the electroless nickel plating process, Te Yuan Chiang et al. has coated the chromium rich nickel layer on the SUS 304 SS and explored the electrical and corrosion behaviour through standard test procedures. He has concluded that the plated samples had healthy ICR and E_{corr} (corrosion potential) value than the unplated bare alloy substrate [5]. Te-Yuan Chiang et al. has developed achromized nickel surface on the steel through the electroplating process and further he has used the pack chromization process in addition for studying the electrical and corrosion property of the sample. He has noticed that the ICR value had shown reasonable improvement and from the polarization plot, it is clearly revealed that the E_{corr} value was quite high for the coated samples [6]. The formation of Ni/SUS 304 SS bi- layer was achieved by Nen-Wen-Pu et al. through the incorporation of the graphene by CVD process. He has observed that there was deposition of graphene layer across the boundary region of Ni/304SS, which has attributed the increase in corrosion resistance value. From the micro graph analysis, the author has concluded that there was uniform distribution of the graphene particles over the grain boundary surface of the Ni-SS interface [7]. Hsiang-Cheng Wang et al. has optimized the process parameters for electroplating the trivalent Cr-C layer on the SS 304 substrate. He has inferred that the sample with constant current density of $10\text{A}/\text{dm}^2$ with varying deposition time has developed a higher corrosion resistance and ICR value in the simulated PEMFC condition [8]. The deposition of CR-N-C multi layer was achieved through the Closed Field Unbalanced Magnetron Sputter Ion Plating (CFUBMSIP) process with 0.1 mm thick SS 316L SS as the substrate. Peiyun Yi et al. has observed that the sample processed with OEM setting of 60% has provided the lower ICR of $2.11\text{ m}\Omega\text{ cm}^2$ under the pressure of 1.4MPa and the corrosion current density was found to be lower having $0.308\text{ }\mu\text{A cm}^2$ in the cell environment [9]. Hsiang-Cheng Wang et al. has performed the electroplating method for coating the Cr-C layer under various current densities. From his research, he has concluded that the corrosion resistance was increased about $9.360 \times 10^{-8}\text{ A}/\text{cm}^2$ by the addition of Cr-C film and with the ICR of $<20\text{m}\Omega\text{ cm}^2$ at $150\text{ N}/\text{cm}^2$. The optimum current density was at $10\text{A}/\text{dm}^2$ [10].

Peiyun Ye et al. has furnished a multi layer coating of Cr-C/a-C: Cr over the SS 316L substrate through the Closed Field Unbalanced Magnetron Sputter Ion Plating (CFUBMSIP) process. From the electrochemical test report, he has revealed that the corrosion potential was 0.236 V vs SCE in the anode side and the corresponding passivation current density was $0.276\text{ }\mu\text{A cm}^2$ at 0.6 V at the cathode end. Both the E_{corr} and ICR were improved as per the DOE targets [11]. Through the Closed Field Unbalanced Magnetron Sputter Ion Plating (CFUBMSIP) technique, Zhiang Wang et al. has postulated the formation of multi layer having C/Al-Cr-N over the SS 316L substrate. The surface conductivity of the multi layer was enhanced through the presence of Cr-N phase and lower Al doping with better corrosion resistance for the SS substrate [12]. N.Mohammad et al. has electrodeposited the lead dioxide over the SS 316L substrate. He has used the plumbite solution of 0.01, 0.03 and 0.05 M Na-F at the ambient temperature for 4 min. The oxygen in the oxide lattice was substituted by the fluorine element and has promoted the electrical conductivity for the sample. The corrosion property was enhanced for the coated substrate than the bare alloy [13]. LvJinlong et al. has investigated the influence of the nano

crystalline SS 316L substrate produced from the cold rolling at liquid nitrogen temperature and followed by electro chemical nitridation process. The author has compared the results of the coated SS with the bare one and concluded that the coated substrate has showed better corrosion potential from the polarization graph [14]. The effect of formation of the nitrogen supersaturated S-phase layer over the SS316L substrate through the active screen plasma nitriding was analysed by Kaijie Lin et al. The author has observed that the presence of S-phase has supported the increment of both the ICR and E_{corr} for the SS substrate [15]. From the comparative study of the ICR value and the corrosion potential of the nickel free SS445, Yang Yu et al. has developed nitride SUS 445 and also Cr-N ion plated SUS 445. Results were compared between SUS 445 – N, Cr-N/ SUS 445 and bare SUS 445 specimens. SUS 445-N specimen showed good electrical conductivity with resistivity of $1.63 \times 10^{-3} \Omega$. The corrosion potential was improved by the presence of super saturated N-phase, Cr-N and Cr_2N phases over the SS surface with less current density [16]. Jie Jin et al. present the importance of the Ti substance that were added on the Cr-N coated stainless steel 316L by the process of Closed Field Unbalance Magnetron Sputter Ion Plantation (CFUBMSIP). From the test data, the Cr-Ti-N-4A has revealed better results than the other samples. The corrosion current density was low of $1 \times 10^{-6} \text{ A/cm}^2$ along with the ICR of $4.7 \text{ m}\Omega \text{ cm}^2$ at compaction force of 1.4 MPa. The contact angle for Cr-Ti-N- 4A was 109° and 99.5° before and after potentiostatic polarization [17]. Lucia Mendizabal et al. have reported the importance of tantalum nitride (TaN_x) coated over the SS316L substrate by High Power Pulsed Magnetron Sputtering (HPPMS). Electro chemical tests were performed under various cathodic potential ($0.8 V_{\text{SHE}}$, $1 V_{\text{SHE}}$, and $1.4 V_{\text{SHE}}$), pH values of (pH 3 & 6) and test timing of (5 and 180 min). All the samples had shown the current density in the range of $1\text{--}15 \mu \text{ A cm}^{-2}$. The tantalum coated SS 316L which was polarized at $0.8 V_{\text{SHE}}$ showed the lower ICR values around $25\text{--}37 \text{ m}\Omega \text{ cm}^2$ than the nitride Ta substrate. The author studied the barrier oxide layer characteristics through Auger Electron Spectroscopy (AES), as these layer influence the ICR values [18]. Malihe Omrani et al. portrays his research work of coating a double layered Al-N/ Ti-N on SS 316L specimen through plasma focus device. These coatings were developed by the plasma focussing device using the shot number of 5, 10, 15 and 20. The author has compared the bare SS sample and the 5, 10, 15 and 20 shot samples. He has revealed that the ICR value for the 20 shot samples at 200 N/cm^2 was 6 and $120 \text{ m}\Omega \text{ cm}^2$. The corrosion current for 20 shot sample was found to be $7 \mu \text{ A/cm}^2$ and polarization resistance was $251 \Omega \text{ cm}^2$. The author has confessed that the ICR value was declined due to the formation of Ti-Ti₂N and Al-Ti₃N crystalline phases on the SS surface [19]. Jie Jin et al. portray the coating carried out on 304 SS with Cr-Ni-N with different nickel content through the process of Closed Field Unbalanced Magnetron Sputter Ion Plating (CFUBMSIP). The author compared his results with Cr-N, Cr-Ni-N-2a, Cr-Ni-N-4a, Cr-Ni-N -6a, and bare 304 SS substrate. The corrosion potential was favourable to the Cr-N sample with -141.9 mV at cathodic side and 153.8 at anodic environment. The ICR results gave low value of $11 \text{ m}\Omega \text{ cm}^2$ for coated Cr-N SS sample under the compaction pressure of 160 N/cm^2 . The value was detained due to the majority of Cr content. The Cr-Ni-N coated substrate has the lower contact angle of 114° than the other samples [20].

M. Alishahi et al. has successfully deposited nano crystalline multi layer coating of tantalum/tantalumnitrate (Ta/Ta-N) over the SS 316L by the reactive DC magnetron sputtering method. He has compared the ICR value and E_{corr} for the coated and bare sample. He has concluded with his results by stating that the current density for the corrosion test for the coated substrate was $2.84 \times 10^{-2} \mu \text{ A cm}^{-2}$ value. The contact angle was estimated to be around 63° and 92° for bare and coated sample. The ICR value for coated sample was $12 \text{ m}\Omega \text{ cm}^2$ that meets the DOE 2017 target [21]. Yang Yu et al. has studied the electro chemical corrosion property and interfacial contact resistance for low cost nickel free 445 SS through the nitriding process. The comparison was done between bare SUS445, SUS 445-N and SUS 445-AR and he inferred that

the corrosion resistance was high for SUS 445 –N, because structure of the nitride SS445-N has switched over from α -Fe to γ -Fe and electrical resistivity was also low having $1.75 \times 10^{-3} \Omega$ [22]. Shengli Wang created Ti/Ti-Cr-N/ Cr-N multilayer coated 316L SS bipolar plate through arc ion plating technique. He declared that the coated sample showed the ICR value of $4.9 \text{ m } \Omega \text{ cm}^2$ under 150 N/ cm^2 compaction force, due to the presence of Ti layer and Cr-N layer. From the electrochemical corrosion test, the coated sample has showed the least corrosion current density of $0.12 \mu \text{ A cm}^{-2}$ and the corrosion potential was $0.075 \text{ V}_{\text{SHE}}$ than the uncoated SS 316L substrate. The coated sample was found to be superior than the bare sample SS in account of both corrosion and conductivity [23]. Feifei Bi et al. Has performed the analysis of ICR and E_{corr} for the SS 316L sample. He has prepared six different ternary coated sample of Cr-Al-N through the process of Closed Field Unbalanced Magnetron Sputter Ion Plating (CFUBMSIP) with a proper Al composition. Among the six samples, he noticed that S2 showed low ICR value of $5.1 \text{ m } \Omega \text{ cm}^2$ under compaction force of 1.4 MPa . From his potentiodynamic polarization result, the S2 showed a highest corrosion potential of 0.047 V and corrosion current was $-0.79 \mu \text{ A cm}^{-2}$ as per the DOE target. He concluded that the lower incorporation of Al in the Cr-N substrate will propel both the conductivity and corrosion properties [24]. Orsi et al. paper revolves on the Ti-N coated SUS 316 L bipolar plates. The author has focussed mainly on the corrosion test parameter like potential, time duration and pH value of the electrolytic medium. He has coated $1 \mu \text{m}$ thin film Ti-N on the SS 316 L through Closed Field Unbalanced Magnetron Sputter Ion Plating (CFUBMSIP). He has noticed that the ICR value was increased depending up on the potential and duration from 12.9 to $287 \text{ m } \Omega \text{ cm}^2$. The optimal parameters were noticed at $1.4 \text{ V}_{\text{SHE}}$, 1 hour and at 1 M sulphuric acid [25]. ChulKyu Jin et al developed a rubber formed SUS 304 with ternary coating of Ti-N, Cr-N and Ti-N/Cr-N through dc magnetron sputtering technique. From his results, the ICR values was found to be $10.2 \text{ m } \Omega \text{ cm}^2$, 22.4 and $23.5 \text{ m } \Omega \text{ cm}^2$ for Ti-N, Cr-N and Ti-N/ Cr-N under 140 N/cm^2 compaction force. From the corrosion test, he noticed that the current densities were 0.69 , 0.62 and $0.65 \mu \text{ A cm}^{-2}$ and Ti-N coating has the higher value for both ICR and corrosion resistance properties [26]. Zhiyuan Wang et al. work illustrates the multi layered coating of C/Al-Cr-N on the SS 316 L bipolar plate. He predicted the ICR and corrosion potential values by altering the Al/Cr composition. Al_2/Cr_4 sample maintains a good ICR value of $4.88 \text{ m } \Omega \text{ cm}^2$ at 150 N/cm^2 . From the corrosion test, the C/Al-Cr-N coating sample having Al content of 32.24% and $1:1$ ratio of Cr-N has showed the good corrosion potential in both the cathode and anode environments [27]. Min Zhang et al. focused on the estimation of the ICR value and corrosion potential of a multi layered Ti-Mo-N coated SS 316 L bipolar plate through hybrid coating system (Arc Ion Plating with Ti and dc magnetron sputtering method with Mo). He has concluded that the Ti-Mo-N layered coating with Mo content lesser than 26.7% has showed good corrosion resistance than bare SS 316 [28]. Pugal Mani et al. has described the various coatings made on the SS 316 L through cathodic Arc Evaporation System (CAE Balzer Rapid Coating System RCS) coating technique. His coating consisted of mono layer Ti-N, Ti-Al-N and bilayer Ti-N/Ti-Al-N. From his results, the contact angle measurement of Ti-N/Ti-Al-N was higher with 112° than other samples. From the corrosion test, he noticed that the Ti-N coated sample showed better current density value of $0.3 \mu \text{ A cm}^{-2}$, corrosion potential of 0.03 V and protection efficiency of 68.5% [29]. Nguyen Dang Nam et al. designed a multi layered SS 316 L bipolar plate of Ti-N, Ti-Al-N and Ti-Al-Si-N through cathodic arc plasma system. He noticed that Ti-Al-Si-N coated sample made a highest corrosion potential of $140 \text{ m V}_{\text{SCE}}$ and a low corrosion current density of $0.5 \mu \text{ A cm}^{-2}$. The protective efficiency was found around 90% for Ti-Al-Si-N, which was higher than the others. Finally, he concluded that Ti-Al-Si-N sample was more conducting on the cathode side and achieves $< 0.01 \text{ m } \Omega \text{ cm}^2$ of ICR [30]. From the revision of the above furnished journals, it was concluded that the main issues for the metallic bipolar plates especially, stainless steel plate would be sorted out through the proper surface engineering techniques and it can

satisfy the DOE targets with optimal durability in account.

3. CONCLUSIONS

Proton Exchange Membrane Fuel Cell (PEMFC) was acclaimed to be one of the eco-friendly devices of generating power with pollution free factor in the transportation application. Bipolar Plate (BP) is a classical part of the PEMFC and recently properties of the BP have become a keen interest, as these plates should possess many functional properties. Many alternative materials are being evolved to reduce its cost and to attain maximum durability. The main issue for metallic BPPs were the corrosion in acidic environment. The phenomenon of corrosion will occur mostly at the anodic and cathodic region which will disturb the durability of both the fuel cell membrane and BPP. Recent trends were focused mainly on the selection of costless and effective coating techniques and also alternate materials for the BPP for eradicating such corrosive effect in PEMFC environment. Low cost, easily available and high quality based surface coating technology is the current need. From the literature survey, there are more lights on different coatings and surface engineering techniques for SS BP that have been used for tailoring the corrosion behaviour and electrical property, as both the parameters were controversial to each other. General review of various surface coating technologies were discussed in this paper from different researchers. They had altered the surface texture of the substrate for promoting maximum durability for the metallic BP as per the DOE targets. Furthermore, many new composite duplex coatings, conductive polymer coating, and carbon-based coatings are yet to be reviewed.

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