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# Development of Corrosion Resistant Platings for BiPolar Plates of Proton Exchange Membrane (PEM) Fuel Cells – A Review

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## ABSTRACT

Bipolar Plates (BPs) are a key component of the Proton Exchange Membrane (PEM) Fuel Cells with multifunctional character. They uniformly distribute fuel gas and air, conduct electrical current from cell to cell, remove heat from the active area, and prevent leakage of gases and coolant. Bipolar Plates also significantly contribute to the volume, weight and cost of the Proton Exchange Membrane Fuel Cell stacks. Hence, there are vigorous efforts worldwide to find suitable materials for Bipolar Plates. The materials include non-porous graphite, coated metallic sheets, polymer composites, etc. This paper reviews various types of materials, in use and proposed, for Bipolar Plates and critically examines their physical and chemical properties.

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## Introduction

The global climate changes are produced by the greenhouse gases emission such as CO<sub>2</sub>, NO<sub>x</sub> and SO<sub>x</sub> that are ongoing throughout the world pose which progressively have higher demand for replacing today's fossil fuel based on the energy production by less pollutant technologies [1,2]. Among the alternative energies available the proton exchange membrane (PEM) fuel cells are considered to be power transportation for vehicles such as automobiles and buses due to the high power density, relatively quick start-up, low operating temperatures and low greenhouse gas emissions [3]. All the main vehicle manufacturers like General Motors, Ford, Toyota and Peugeot are developing fuel cell cars. Honda launched the first commercial fuel cell car, the FCX Clarity, in the United States market during 2008 summer [4]. Nevertheless, to achieve completely the automotive industry requirements PEM fuel cells have to overcome some intrinsic limitations mainly related to the durability and cost compared to the conventional internal combustion engines [5].

The use of the lightweight metals for the fuel cell bipolar plates are attractive for the automotive use. High corrosion resistance and electrical conductivity requirements for the fuel cell components, however, preclude most uncoated metals from use. Careful selection of the alloy coatings and their constituents reduce or control the electrochemical corrosion potential and the corresponding corrosion rate of the alloy coating. New low-cost alloy coatings are developed for the aluminium that possess the requisite high corrosion resistance and high electrical conductivity. The current development efforts include novel electrolytic alloys and conductive

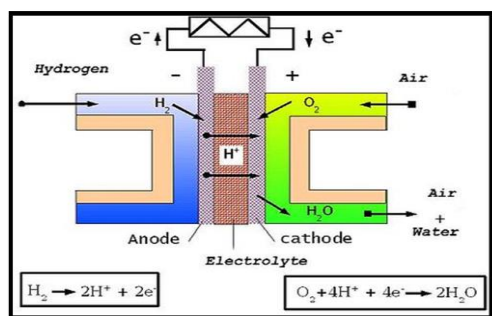
polymer coatings for aluminium to achieve the desired fuel cell component lifetime goals.

## Development of the Corrosion Resistant Platings for the Fuel Cell Bipolar Plates

Proton Exchange Membrane Fuel Cells are of prime interest in transportation applications due to their relatively high efficiency and low pollutant emissions. Bipolar Plates are the key components of these devices as they account for significant fractions of their weight and cost. Metallic materials have advantages over the graphite-based plates, because of their higher mechanical strength and better electrical conductivity. However, corrosion resistance is a major concern that remains to be solved as metals develop oxide layers that increase electrical resistivity, thus lowering the fuel cell efficiency. The paper aims to present the main results found in recent literature about the corrosion performance of the metallic bipolar plates.

## Fuel cell

A fuel cell is a device that converts the chemical energy from a fuel into electricity through a chemical reaction with oxygen or another oxidizing agent.[1] Or in other words as follows. Fuel Cells are Electro Chemical Devices consisting of an Ion Exchanging Solution, Liquid or Solid in contact with two electrodes capable of converting directly any consumable Fuels to Electrical Energy through the performance of a Chemical Reaction.



Hydrogen is the most common fuel, but hydrocarbons such as natural gas and alcohols like methanol are sometimes used. Fuel cells are different from the Batteries. In that they require a constant source of fuel and oxygen to run, but they can produce electricity continually for as long as these inputs are supplied.

### Fuel Cells – Relevance and Importance

‘What’s so great about Fuel Cells? ‘Should be a question synonymous with ‘What’s so great about Pollution, Changing the climate or running out of oil, natural gas and coal?’ When it is head into the new millennium, it is the time to put renewable energy and planet-friendly technology at the top of the priorities. Dawn of this new millennium witnesses a triumphal technological march centered on the Fuel Cells! ‘Clean Energy’ is a technological edict that is echoing around the global.

Fuelled by the concerns about the environmental pollution, energy security and climate change and the notion of a ‘Clean Technology’s moving beyond the realm of scientist and engineers and into the lexicon of the political and business leaders. Energy is the lifeblood of the global economy. Getting and sharing information and the producing things require energy. Throughout the industrial age and into the information age, energy has served as the foundation for the mankind’s progress.

Technology is the prime propellant of the growth, modernization, progress, prosperity, power and prestige. There is no escape from the accelerating effect and the massive impact of the technology is in the people’s outlook, life styles-social and cultural-and economic structure.

If greater capacity of a nation to generating, transferring and utilizing technology, the faster is the nation’s growth. The goal is the development, the tool is technology and the path is Science. Hence, today Science and technologies are the chariot wheels of the development process.

Few technologies have the potential to change the World in to the better as the Fuel Cell, which offers a potentially non-polluting and renewable way to generate electricity. Fuel Cells are efficient, clean, safe and reliable. They attract increasing commercial interest for both the automotive and stationary power generation. The advent of the Fuel cells is powering and energy revolution.

Hydrogen represents an alternative source for energy carrier to the oil based economy. Fuel cells are the harness hydrogen for the energy production and thus lead to the transition. To exploit fully the capabilities and the operational advantages, the research and development of the fuel cells have to confront innumerable challenges and have to wait for a variety of technological breakthroughs. Fuel Cells have the potential to satisfy the future energy needs.

Air and space power professionals presently review the ongoing conflicts over energy and fossil-fuel resources with the intention of making this transformation potential a reality as well as to understand the promise and limitations of fuel cell technologies, so as to take advantage of the transformation.

### The attractive features of the Fuel Cells are,

- ✓ They are simple with the possibility of the direct energy conversion
- ✓ They show efficiencies greater than the existing fuel burning engines and also work with a flat efficiency curve even at the part loads.
- ✓ They are silent without any rotating/moving components.
- ✓ Fuel cell power plants are constructed in the modular mode to match load and increase reliability.
- ✓ Fuel and size flexibility is possible.
- ✓ Fuel cells are exploited for the Combined Heat and Power (CHP) generation with more electricity (40-45% and 35% - 40% heat)
- ✓ They do not generate SO<sub>x</sub> and NO<sub>x</sub> emissions.

How fast might the energy system evolve towards Hydrogen? Energy transitions are driven by growing energy demands, local scarcities and the continual search for more abundant and accessible energy sources. *The 19th century was considered to be the age of the INTERNAL COMBUSTION ENGINE and 21st Century may well come to be recognized as the age of the FUEL CELLS.*

### What are Fuel Cells?

Fuel Cells are electro chemical devices consisting of an electrolyte, an ion containing solution, liquid or solid in contact with two electrodes (anode where the Oxidation of the Fuel takes place and a cathode where the reduction of Oxygen occurs) capable of converting directly any consumable fuels to electrical energy through the performance of a chemical reaction.

In the production of electricity through this mode the by-products are Heat, Carbon Di-Oxide and Water and it can be considered to be avoiding any of the environmental side effects. The electro chemical process in the fuel cell is efficient in converting nearly 80% of the chemical energy of the fuel into electricity in contrast to the thermal power plants which give efficiency around 40%. In the structure, Fuel Cells are similar to the Batteries. The difference, in batteries the chemical energy is stored and need to be recharged or replenished, while in the fuel cells the fuel is constantly fed to the cell so that consistent generation of electricity is ensured.

Fuel cells are constructed in the modular form and hence fuel cell power plants are erected to provide electricity for a number of applications ranging from electric vehicles to large grid is connected for the utility power plants and for the stationary power for communities. A single cell normally produces a voltage around 1.0 V. To obtain higher voltages the cells have to be connected in series to form a stack. The heat rejected in the process is used for different on-site thermal consumption which makes the fuel cells suitable for the combined heat and power generation for building and industry.

## Fuel Cells: How do they differ from Batteries?

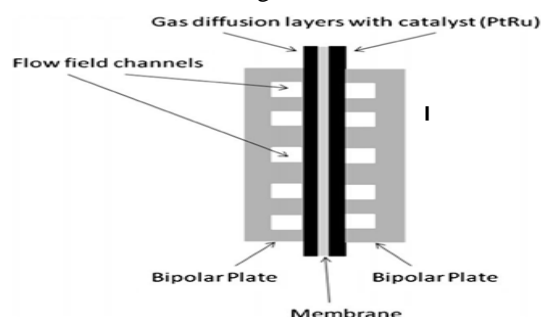
Fuel Cells have similar components and characteristics of a battery, but differ from them in several ways. A battery is an energy storage device and the extent of storage is determined by the amount of the chemical reactant stored within the battery itself. In the primary battery the maximum energy available is determined by the amount of the chemical reactant stored and will cease to produce electrical energy when the chemical reactants are consumed or discharged.

However, in a secondary battery, the recharging involves the transferring energy from an external source and regenerates the reactants. A fuel cell however, as stated above is the energy as long as the fuel and oxidant are supplied to the electrodes. General wear and tear, electrochemical corrosion, the material stability and malfunctioning of the components may limit the life of fuel cells.

Depending on the nature of the ionic species involved in the electrolytic cell, the direction of the transport of the species can differ. Similarly the site of water (oxidized product) formation and removal can be different. The fuel or oxidant flows over the surface of anode and cathode and generate electrical energy by the electrochemical oxidation of the fuel (in this example hydrogen) and electrical reduction of the oxidant (usually oxygen).

*The electrolyte not only helps in the transportation of the dissolved reactants to the electrodes but also conducts ionic charge between the electrodes and thus completes the electric circuit. The electrolyte can also acts as a barrier to prevent fuel and oxidant gas streams to mix with each other, which will reduce the efficiency of the fuel cell.* The functions of the electrodes in a fuel cell can be listed as follows.

The main components of a PEM fuel cell structure are bipolar plates and the membrane electrode assembly (MEA). The MEA comprises the proton exchange membrane, gas diffusion layer (GDL) and a catalyst layer. A schematic view of such structure is shown in Fig. 1.



In a fuel cell stack the bipolar plates are key elements as they account for large fractions of the total weight, volume and cost of the stack. According to Tsuchiya and Kobayashi [6] bipolar plates may reach up to 80% of the total weight and 45% of the total cost in a PEM fuel cell stack. More recent data show that the relative cost has fallen to about 25% of the stack, yet a very significant amount.

Furthermore these components play vital functions in the stack to carry electric current away from each cell, to distribute the fuel and oxidant homogeneously within individual cells, to separate individual cells and to facilitate

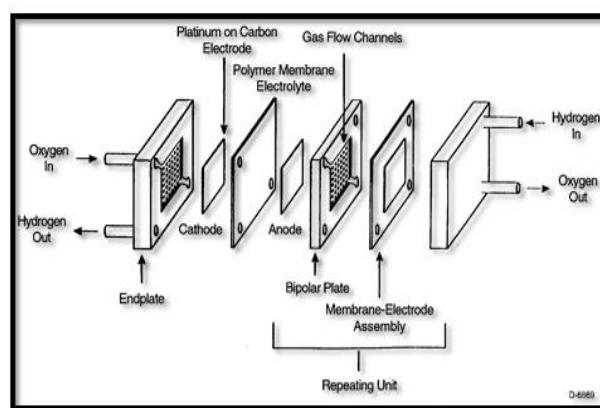
the water management within the cell [8]. To perform such a number of functions a variety of materials are proposed to manufacture bipolar plates. The main properties that any material must present to be used in these devices are well established by the United States Department of Energy (DOE) as shown in the Table 1. The criteria shown in the table are must-pass technical requirements that any material should be capable of achieving in order to be as considered apt bipolar plate material.

The earlier traditional option was the use of the non-porous graphite plates due to their intrinsic high electrical conductivity and chemical stability in the PEM fuel cell in an aggressive environment. Although the performance of the graphite plates are suitable related to the properties they brittle and lack mechanical resistance. Another drawback is the need for machining the flow field channels which raises the manufacturing cost making it prohibitive for mass production [9].

Manufacturers of the next generation vehicles are targeting the fuel cell power sources due to their higher efficiency operation and reduced emissions than the present-day combustion processes. One of the leading candidate fuel cell systems for transportation application is the hydrogen/air proton exchange membrane fuel cell (PEMFC). Fuel cells for the application requires 30 to 50 kW levels, depending on the chosen vehicle and load-levelling requirements. Attainment of power levels is accomplished by stacking individual cells or membrane electrode assemblies (MEAs), each separated by a bipolar plate.

The bipolar plates serve three purposes:

- 1) Separation of the fuel and oxidant gases,
- 2) Rigidity for the MEA, and
- 3) Electron flow through the fuel cell stack.



**Figure 1.- PEM Fuel Cell Components**

Figure 1 illustrates the components of a PEM fuel cell stack. Traditionally, these plates are fabricated from the graphite, owing to its chemical nobility, and high electrical and thermal conductivity.

The major impediments to the commercialization of PEMFCs for the automotive applications are the cost, weight, and volume of the bipolar plates.

State-of-the-art PEMFC stacks contain graphite bipolar plates that are either machined or moulded with a flow field arrangement for gas flow to the electrodes. To increase the

structural strength and minimize gas permeation, the plates are usually relatively thick, allowing the gas channels on the both sides of the plate. A typical 60-cell stack, therefore, becomes excessively large and heavy.

Bipolar plates constitute the backbone of a hydrogen fuel cell power stack, conduct current between cells, facilitate water and thermal management through the cell, and provide conduits for the reactant gases namely hydrogen and oxygen. In the polymer electrolyte membrane (PEM) hydrogen fuel cell design, bipolar plates are fabricated in the mass production and they must be made of materials with the excellent manufacturability and suitable for the cost-effective high volume automated production systems.

Currently, graphite composites are considered as the standard material for PEM bipolar plates because of its low surface contact resistance and high corrosion resistance. Unfortunately, graphite and graphite composites are classified as brittle and permeable to the gases with poor cost effectiveness for high volume manufacturing processes relative to the metals such as aluminum, stainless steel, nickel, titanium, etc. Since the durability and cost represent two main challenges hindering the fuel technology from penetrating the energy market and competing with other energy systems, considerable attention is recently given to the metallic bipolar plates for their particular suitability to the transportation applications.

Metals enjoy higher mechanical strength, better durability to shocks and vibration, no permeability, and much superior manufacturability and cost effectiveness when compared to the carbon-based materials, namely carbon-carbon and carbon-polymer composites. However, the main handicap of the metals is the lack of ability to combat corrosion in the harsh acidic and humid environment inside the PEM fuel cell without forming oxidants, passive layers, and metal ions that cause considerable power degradation. Considerable attempts are made using the noble metals, stainless steel and various coated materials with nitride- and carbide-based alloys to improve the corrosion resistance of the metals used without sacrificing surface contact resistance and maintaining cost effectiveness.

Gold-Plated titanium and niobium are the materials used by the General Electric in the 1960s [1] that were later replaced by graphite composites to reduce cost and weight. In recent years, due to lack of graphite durability under mechanical shocks and vibration combined with cost effectiveness concerns of its high volume manufacturability, considerable research work is currently underway to develop metallic bipolar plates with high corrosion resistance, low surface contact resistance, and inexpensive mass production.

Various types of metals and alloys are currently under the testing and evaluation by the researchers working in the field of PEM fuel cells to develop the bipolar plates that possess the combined merits of graphite and metals. The ideal characteristics of a bipolar plate's material is high corrosion resistance and low surface contact resistance, like graphite, and high mechanical strength, no permeability to reactant gases and no brittleness like metals such as stainless steel, aluminium, titanium, etc.

The main challenge however is that corrosion resistant metal bipolar plates develop a passivating oxide layer on the surface that does protect the bulk metal from progression of corrosion, but also cause an undesirable effect of a high surface contact resistance. This causes the dissipation of some electric energy into heat and a reduction in the overall

efficiency of the fuel cell power stack. The key characteristics of bipolar plates material that are suitable for transportation applications are as follows:

- high corrosion resistance with corrosion current at 0.1 V and  $H_2$  purge  $< 16 \text{ A cm}^{-2}$ ;
- high corrosion resistance with corrosion current at 0.6 V and air purge  $< 16 \text{ A cm}^{-2}$ ;
- Interfacial contact resistance (ICR) @ 140 N  $\text{cm}^{-2} = 20 \text{ m}\Omega \text{ cm}^2$ ;
- does not dissolve and produce metal ions;
- possess steady low Ohmic resistance throughout the operation;
- high surface tension with water contact angle close to  $90^\circ \text{C}$ , i.e. high dehydration;
- light weight;
- high mechanical strength  $< 200 \text{ N m}^{-2}$ ;
- High volume cost-effective manufacturability: US\$ 10  $\text{kW}^{-1}$ .

Alternatives to graphite and including conductive polymers are explored, but with only limited success. The use of the plated and non-plated metal bipolar plates has received attention recently due to the simplicity of stamping a flow field into the metal. The thin nature of the metal substrate allows for smaller stack designs with reduced weight. The Table 1 lists the benefits of the metallic bipolar plates, and the Figure 2 illustrates the effect for an entire 33kW PEMFC stack. From a cost perspective, inexpensive metals such as stainless steel and aluminium could easily be processed into bipolar plates. Titanium has been suggested as an alternative for the PEMFC bipolar plate material, but are better suited for aerospace applications rather than for low-cost automotive use.

***One of the unfortunate aspects of the metallic bipolar plate materials is corrosion or oxide layer formation that lowers the electrical conductivity and severely reduces fuel cell performance.*** 316 stainless steel is successfully used as a bipolar plate material by avoiding any contact with the ion-exchange membrane. However, Mallant et al.<sup>1</sup> found that PEMFCs using stainless steel bipolar plates showed about a 10% degradation in fuel cell performance over 1000 hours. Assuming, at the best, a linear degradation, at the least 50% of the fuel cell performance (power) would be lost over 5000 hours. Unfortunately, current design goals require greater than 5000 operating hours, as well as numerous start- stop cycles.

A few researchers have pursued Plating processes to protect the metal bipolar plates. Aluminum is plated with the conductive materials by Chemical Vapor Deposition (CVD) processes and ion sputtering. These techniques require high processing temperatures; Low-temperature plasma spraying is investigated, but results in severe pitting of the aluminium substrate. Both the techniques require high capital equipment costs. Therefore, an effective, low-cost approach is needed to fabricate corrosion resistant Platings on the aluminum bipolar plates. Below the Table summarizes the anticipated cost benefit of plated metal.

Alternatives to pure graphite plates are composite bipolar plates based on the mixture of polymers and graphite particles. This class of materials allows mass production at a reasonable cost using the manufacturing processes such as **injection moulding for thermoplastics or BMC for thermo sets.**

There are several examples of graphite-based composite

Bipolar plates using Polypropylene (PP) and Poly Phenylene Sulphide (PPS), Phenolic and Vinyl Ester resins as matrices [10–14]. The polymer matrix gives flexibility to the bipolar plate improving its mechanical strength. The chemical stability is also not badly affected by the incorporation of polymer in graphite. On the other hand, electrical conductivity is proportionally diminished since polymers are insulating materials. Thus it is mandatory to formulate a composite bipolar plate with carefulness to attain mechanical performance without sacrifice electrical conductivity.

#### Performance Requirement for the Proton Exchange

##### Membrane Fuel Cell (PEM) Bi-Polar Plates

Property	Unit	Value
Flexural strength – ASTM D790	MPa	>59
Electrical conductivity	S cm <sup>-1</sup>	>100
Corrosion rate	mA cm <sup>-2</sup>	<1
Contact resistance	mΩ cm <sup>2</sup>	<20
Hydrogen permeability	cm <sup>3</sup> (cm <sup>2</sup> s) <sup>-1</sup>	<2.10 <sup>-6</sup>
Mass	kg/kW	<1
Density – ASTM D792	g cm <sup>-3</sup>	<5
Thermal conductivity	W (m K) <sup>-1</sup>	>10
Impact resistance	J m <sup>-1</sup>	>40,5
(unnotched) ASTM D-256		

In spite of all the advantages of the graphite-based composite bipolar plates regarding to the low weight, high

production and chemical stability, if one compares their overall performance with the metal bipolar plates two major drawbacks become evident, that is, their lower mechanical resistance and electrical conductivity. Considering especially transportation applications metal bipolar plates are more resistant to the mechanical shocks and vibrations that could lead to the cracking and leaking of the reactant gases. Cunningham [15] presented data showing that the electrical conductivity of the metal bipolar plates may reach up to 1000 times that of the composite ones. In addition, they present easy manufacturability at low cost which increases their competitiveness in the fuel cell market [16]. However, a significant handicap that may decrease the metal bipolar plates' performance is the susceptibility to the corrosion in the acid and humid environment of PEM fuel cells. Metals operating in the fuel cell with a pH of 2–4 and temperatures around 80°C may suffer dissolution. The ions leached may poison the membrane electrode assembly (MEA), decreasing the power output of the fuel cell [17,18]. Furthermore, passive layers formed during operation increase the electrical resistivity of metal bipolar plates. Consequently, the fuel cell efficiency is also negatively affected due to the raising of the interfacial contact resistance as the oxide layer grows. These effects offset the advantage of the high electrical conductivity [19].

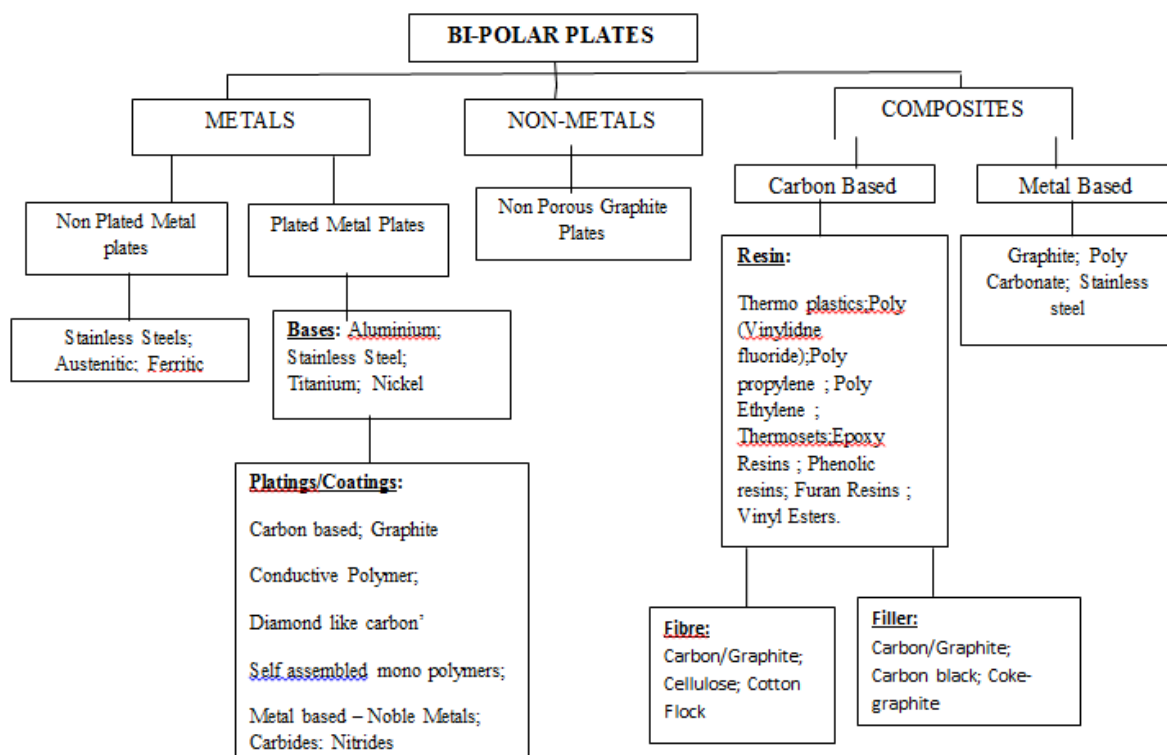


Fig 2. Classification of materials for BPs used in PEM fuel cells.

## Design Criteria for Bi-Polar Plate Materials

Sl.No	Material Selection Criteria	Limit
1	Chemical Compatibility	Anode face must not produce disruptive hydride layer. Cathode face must not passivate and become non conductive.
2	Corrosion	Corrosion rate < 0.016 mA cm <sup>-2</sup>
3	Cost	Material + Fabrication, US\$ 0.0045 cm <sup>-2</sup>
4	Density	< 5 g cm <sup>3</sup>
5	Dissolution	Minimization of Dissolution(For Metallic Plates)
6	Electronic Conductivity	Plate Resistance < 0.01 ohmcm <sup>3</sup>
7	Gas diffusivity/impermeability	Maximum average gas permeability < 1.0X10 <sup>-4</sup> cms <sup>-1</sup> cm <sup>-2</sup>
8	Manufacturability	Must be low with high yield
9	Recyclable	Material can be recycled during vehicle service, following a vehicle accident or when vehicle is retired.
10	Recycled	Made from recycled material
11	Stack Volume/kW	Volume < 11/kW
12	Strength	Compressive Strength 22lb-in <sup>-2</sup>
13	Surface Finish	>50 micro meter
14	Thermal Conductivity	Material should be able to remove heat effectively.
15	Tolerance	>0.05 mm

The problems outlined above overcome or minimize by protecting the metal bipolar plates from the corrosive fuel cell operating conditions with Platings [20]. A wide variety of the alternatives have been proposed in the research works towards the objective. The paper aims to present major research topics and results in the corrosion protection and characterization of the metal bipolar plates for PEM fuel cells. Different Plating methods and substrate materials are addressed giving a comprehensive overview on the subject.

## Mass and Volume Comparisons of Bipolar Plate Materials

Bipolar Plate Type*	Features	Mass	Mass Reduction	Thickness	Thickness Reduction
Graphite	0.76 mm deep grooves 0.76 mm wide channels 0.76 mm wide lands Minimum thickness: 0.76 mm Maximum thickness: 2.29 mm	138 g	-- -	2.29 mm	-- -
Gold Plated Aluminium	0.25 mm thick aluminium 2 µm thick coating 0.76 mm deep grooves 0.76 mm wide channels 0.76 mm wide lands	55 g	60%	1 mm	56%
*Assumes 250 cm <sup>2</sup> active area with a 2 cm border					

## Bipolar Plate Materials Comparison

Material	Conductivity (S/cm)	Cost (\$/kg)	Pros	Cons
Graphite	7	75	<ul style="list-style-type: none"> <li>Immune to corrosion</li> <li>Satisfactory electrical conduction</li> <li>Mouldable</li> </ul>	<ul style="list-style-type: none"> <li>Heavy</li> <li>Expensive</li> <li>Milled flow paths</li> </ul>
Conductive Plastics	0.03 to 3.2	5 to 30	<ul style="list-style-type: none"> <li>Injection moulding</li> </ul>	<ul style="list-style-type: none"> <li>Low conductivity</li> <li>Brittle</li> </ul>
Coated Metal	5.5 x 10 <sup>5</sup>	7	<ul style="list-style-type: none"> <li>Thin</li> <li>Lightweight</li> <li>Stampable</li> </ul>	<ul style="list-style-type: none"> <li>Non-adherent coatings</li> <li>Corrosion</li> </ul>

To demonstrate feasibility of replacing the graphite with aluminum for PEMFC bipolar plates required

Four steps:

- 1) Identification of a corrosion-resistant coating,
- 2) Confirmation of mechanical integrity of coating over the temperature range of PEMFC operation,
- 3) Fabrication of the bipolar plates, and
- 4) Operating performance evaluation and comparison with graphite. Experiments were conducted to meet these demonstration goals.

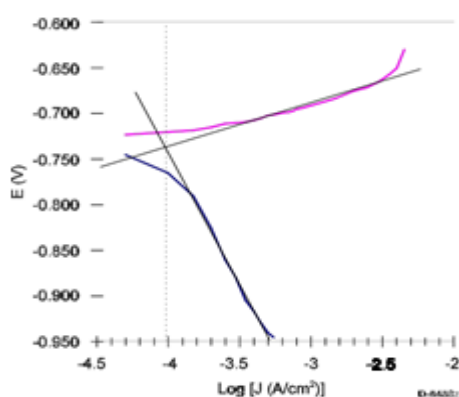


### Corrosion Testing

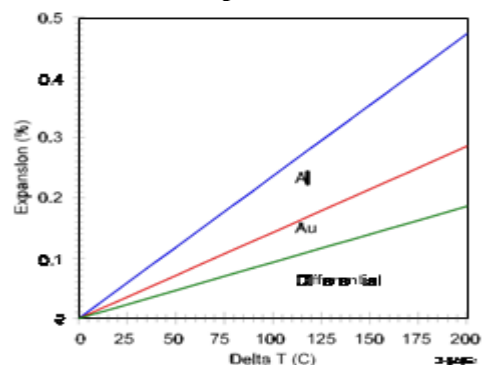
High corrosion resistance is necessary condition for the PEMFC bipolar plate coatings since the operating environment is extremely corrosive. Typical assembly layouts require intimate contact of the bipolar plate and carbon electrodes on a solid polymer electrolyte. An abundant proton source coupled with large potential fields provides the pathway for the electrochemical corrosion. There are numerous corrosion resistant metallic and alloy coatings commercially available, which can be electrodeposited. However, few are adaptable to the bipolar plate Platings since the mechanism for the corrosion protection is the rapid formation of a stable surface oxide (passivation) which prevents the further corrosion. Unfortunately, such oxides are usually not very conductive which departs from the high electrical conductivity requirement of PEMFC bipolar plates.

Towards the goal of identifying the candidate materials for Plating aluminum bipolar plates, several different metals and alloys are evaluated for the corrosion resistance. Faraday's law normalized to time and exposed surface area of the material gives the corrosion rate:

$$R_{\text{corr}} = (ja) / (nF)$$



Differential Thermal Expansion of Gold on Aluminium.



Prior to the corrosion testing of the electrodeposits, pure foils of each material were obtained and the corrosion was tested. This screening process significantly reduced the time and expense of preparing electrodeposits of each material in-house. Following the screening tests, confirmation experiments were conducted on the copper and nickel platings plated on the aluminium substrates. The corrosion rates for these electrolytic coatings matched the corrosion rates for the pure foils. Finally, electrodeposited coatings of the candidate corrosion resistant materials were prepared and the corrosion tested. The results of these corrosion tests are summarized in the Table 3.

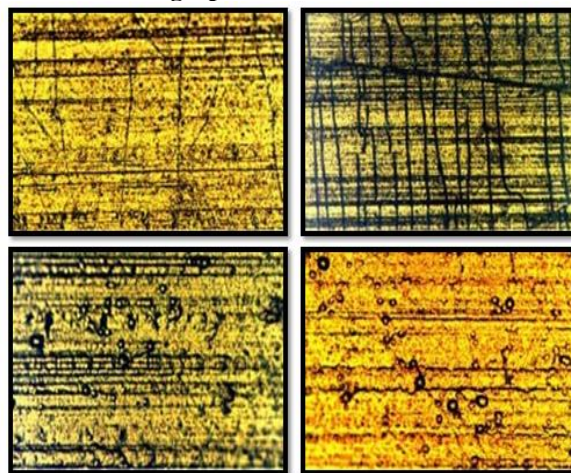
As expected, materials such as aluminium, copper, nickel, and tin are very susceptible to the electrochemical corrosion in the acidic solutions, typical of PEMFC operating conditions. However, materials such as gold and phosphorous nickel show very high resistance to the electrochemical corrosion,

comparable to the graphite, the traditional bipolar plate material. These materials are selected for further study. It is important to note that while thick platings of gold is prohibitively expensive for the vehicle PEMFCs, gold plating thickness of less than 2  $\mu\text{m}$  provides adequate corrosion protection and were re-evaluated for the economic feasibility.

### Corrosion Data for Pure Metals and Commercial Alloy Platings

Materi	Corrosion Rate ( $\mu\text{m}/\text{year}$ )
Aluminum	~ 250
Copper	> 500
Gold	< 15
Graphite	< 15
Nickel	> 1000
Silver	< 15
Tin	> 10000
Titanium	< 100
Tungsten	< 100
Zinc (zincate)	> 2000
Gold-nickel	
Phosphorous Copper (electrolytic)	~ 500
Phosphorous Nickel	< 30
Stainless Steel (316)	< 100
Tin-Zinc	---
Zinc-Nickel	---

### Photo Micrographs of Gold-Plated Aluminium



Little or no micro cracking of the gold plating is seen in the above Figure. It suggests that the micro cracking of the gold is a result of internal stress. The Figure 6(d) shows the surface following thermal cycle tests. Here, only a few micro cracks are present on the surface of the gold plating. A comparison of Figures 6(b) and 6(d) suggest that the plating failure is preventable by controlling the CTE mismatch.

Following the thermal cycle tests on these coupons, corrosion tests are performed to confirm the results of the visual inspection. The Table 4 summarizes the corrosion test data for the 2  $\mu\text{m}$  thick gold deposits. It is immediately obvious from these data that the corrosion measured is that of the underlying material, and not the gold outer layer. This is likely due to the micro cracks in the gold outer layer which is observed during the optical inspection. Such cracks in the gold coating are apparently sufficient to allow the corrosion of the preceding layer. The hypothesis can be confirmed by comparing the as-plated gold over the aluminium sample (6a) corrosion rate with that of the zincates (from Table 3), which was the processing step used to prepare the aluminium for subsequent electro deposition steps.

The corrosion rates are very similar. The same comparison can be made for the As-plated gold over the nickel over the copper and over the aluminium sample (6c). The corrosion rate matches that of nickel which is the coating layer preceding gold. The lower corrosion rates for the thermal cycled samples, despite the obvious mechanical degradation observed during the optical inspection is likely a result of an oxide formation on the coating layer below the gold. The underlying surfaces of zinc (6a) and nickel (6c) passivate very quickly at thermal cycling temperatures near 300°C, thus providing additional corrosion protection. As previously discussed, oxide formation is the mechanism for corrosion protection for many metals and alloys.

### 2.1.1 Bare substrates

#### Stainless Steel

It is generally agreed that stainless steels are prone to the chemical attack in the PEM fuel cell environment. Their corrosion products may poison the catalysts in the polymeric membrane and the oxide layer grown on the metal surface increases the interfacial contact resistance decreasing the power output of the fuel cell [21]. The criterion to select the stainless steels for the bipolar plate applications sharply depends on the chemical composition of the material as the nature and content alloying the elements strongly to influence the composition of the passive film formed on the metal surface which in turn affects its overall corrosion resistance. Hornung and Kappelt [22] used the pitting resistance equivalent ( $\text{PRE} = \frac{1}{4} \% \text{Cr} + 3.3x \% \text{Mo} + 30x \% \text{N}$ ) to rank and select the different iron-based materials for the bipolar plates. The authors compared the performance of these alloys with the nickel-based and Gold-Plated iron-based alloys. The alloys composition was not given in the text. Only the Gold-Plated alloy was presented in the suitable contact resistance. Kim et al. [23] have also investigated stainless steels contact resistance for the eleven different alloys are exposed to the sulphuric acid solution to simulate the PEM fuel cell environment. They showed extensive results joining PRE numbers with the transpassive potentials and contact electric resistance (CER) values. Chromium and molybdenum contents were found to be decisive for decreasing the CER values.

The work of Wang et al. [17] compared the electrochemical behaviour of 316L, 317L, 349 and 904L stainless steel grades in simulating the fuel cell anode and cathode environments. The authors related the Cr content with the corrosion and interfacial contact resistances of the material. The results are pointed towards the following performance order  $349 > 904L > 317L > 316L$ . The order shows that the higher Cr content and the higher corrosion resistance. It was verified that the thickness of the passive layer on the 349 grade was constant with time when the material was polarized at 0.6V during 30 min. Obviously this period is too short for a definite and accurate conclusion. However the results clearly allowed stating that the 349 is the best candidate for the bipolar plate applications. An important fact, not mentioned in the Wang's paper is the influence of both nickel and molybdenum contents on the contact resistance of the passive films formed on the stainless steels.

### 2.1.2 Surface Treatments

Lee et al. [36,37] suggested an electrochemical surface treatment on the stainless steel bipolar plates to improve their corrosion resistance and minimize the formation of a thick

oxide layer that increases the interfacial contact resistance during the operation. The results showed an increase in the breakdown potential and a decrease in the corrosion current density after the treatment. They described this superior behaviour to the Cr enrichment of the oxide layer as verified using the electron spectroscopy analysis. No details on the composition of the electrolyte used to perform the electrochemical treatment were given in the text. Cho et al. [38] evaluated the corrosion resistance of the chromized 316L in the PEM fuel cell simulating conditions. The chromium layer was produced by a pack cementation process. The treatment was effective with regard to the improvement of the corrosion resistance of bare 316L. The lowest values of the current density as shown by the potentiodynamic and potentiostatic tests were obtained for a 2.5h period of treatment. These results were due to a Cr-rich layer with few defects. In a more recent investigation [39], the same authors used another pack cementation process to produce the corrosion resistant chromized 316L stainless steel bipolar plates. They varied the Cr content in the powder mix used for the Cr pack cementation and the heating time during the chromizing treatment. The microstructure of the chromized layer was found to be strongly related to these process parameters.

### Platings

The use of the corrosion resistant and high conductive coatings is an another surface modification technique envisaged as a solution to the lack of corrosion resistance of the metal bipolar plates. A physical vapour deposition (PVD) method was used by Li et al. [55] to deposit titanium nitride (TiN) Plating on the 316L stainless steel bipolar plate as an alternative to protect the materials against the corrosive electrolyte of PEM fuel cells. Potentiodynamic polarization curves showed that the corrosion current density and passive current density of 316L were both decreased after the deposition of TiN, while the corrosion potential was shifted to the nobler values. Furthermore, no signs of pitting were identified in the coated 316L specimens while in the bare specimens the polarization curves presented a breakdown potential at around 700m V SCE. Despite the Promising electrochemical performance of TiN-Plated 316L one concern was pointed out by the authors. Using the SEM they found that the coating was lost on the small areas on the material surface after 1000 h of immersion under cathodic conditions and after 240 h of immersion under anodic conditions. Consequently larger substrate areas may be exposed to the electrolyte with the increase of immersion time. This behaviour has been ascribed to the presence of intrinsic defects on the coating layer such as pinholes and macro particles that are inherent to PVD techniques [56]. A SEM micrograph of a PVD TiN-Plated 316LSS is presented in the Fig. 2. The defects of PVD film are clearly seen in this image.

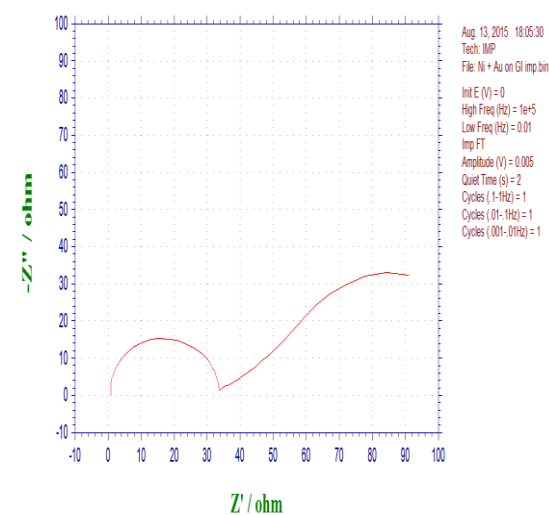
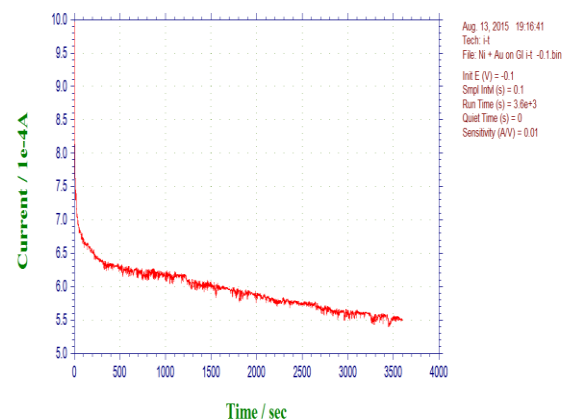
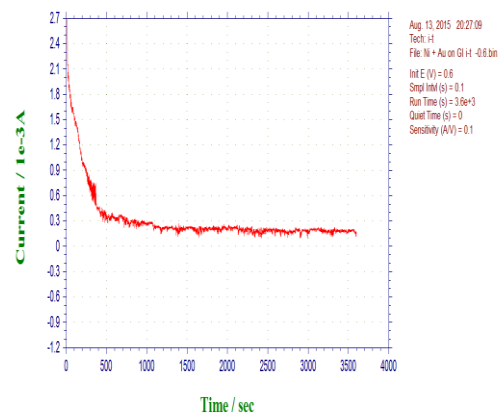
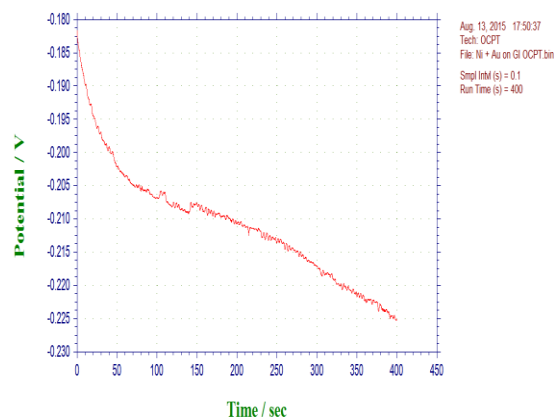
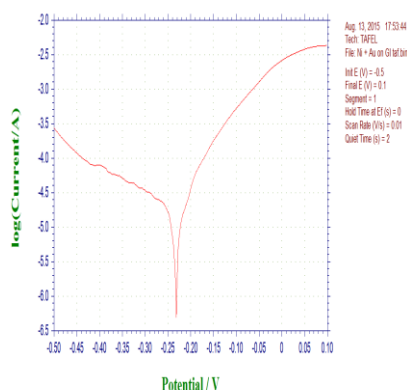
In order to avoid this effect Li et al. suggested changes in the deposition parameters to improve the coating quality. Cho et al. [57] have also found that TiN coating on 316 stainless steel substrate provided good electrochemical performance in the fuel cell environment. However, they identified a coating degradation phenomenon responsible for increasing the charge transfer resistance and, in addition, lowering the ionic conductivity of the membrane due to poisoning by Fe, Cr, Ni and Ti ions released by the 316 substrate and TiN Plating. Wang and Northwood [58] conducted potentiodynamic tests with TiN-Plated 316L stainless steel specimens and

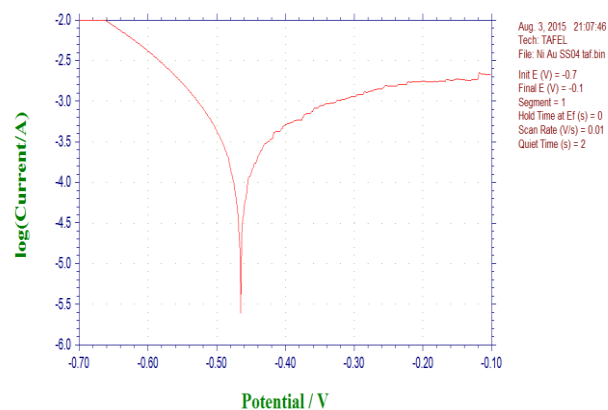
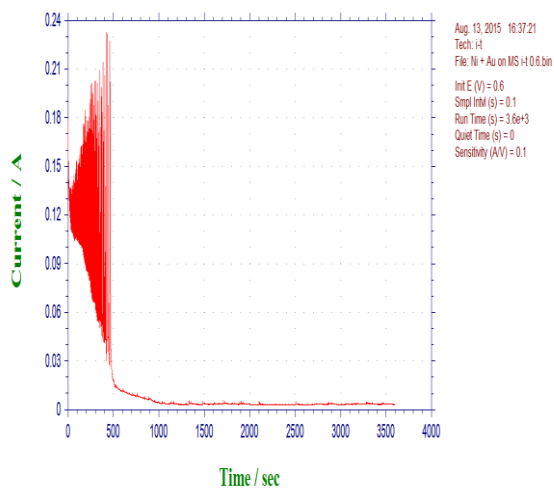
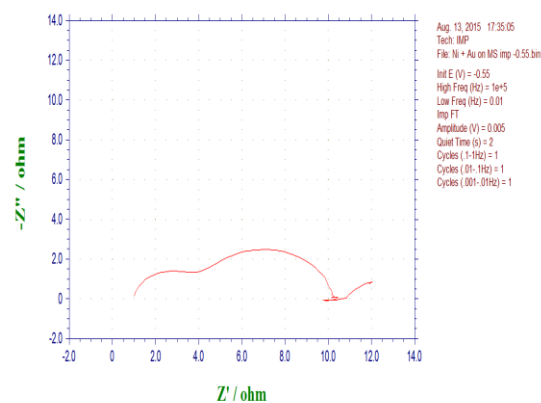
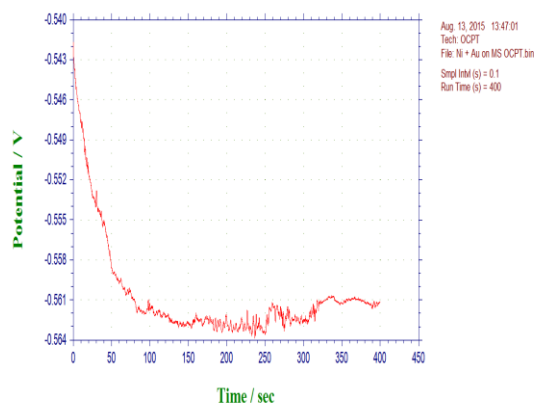
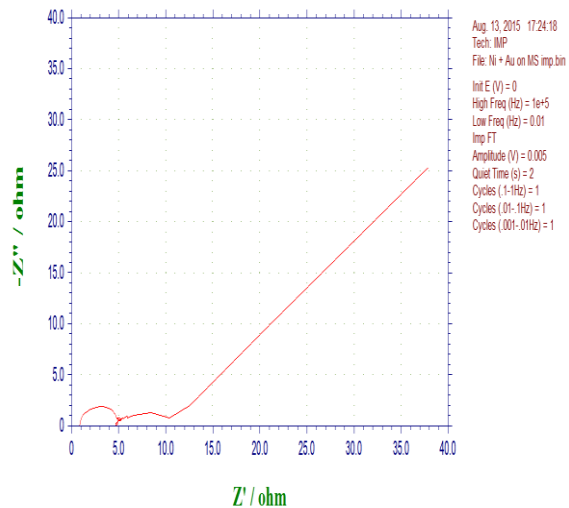
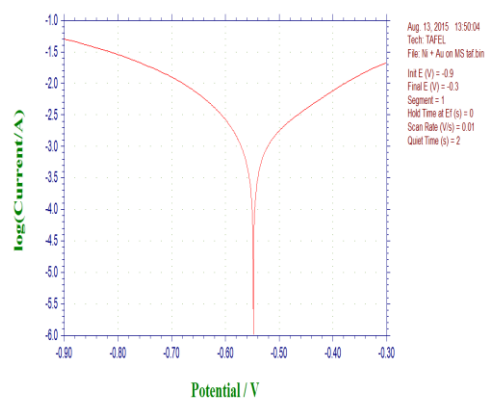
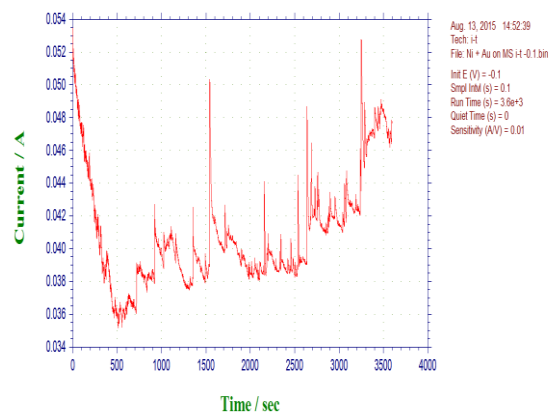
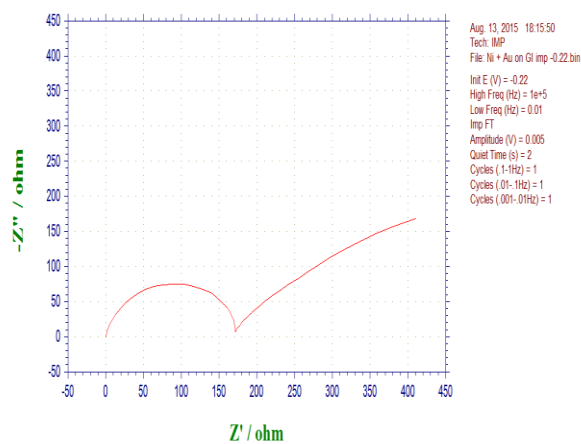


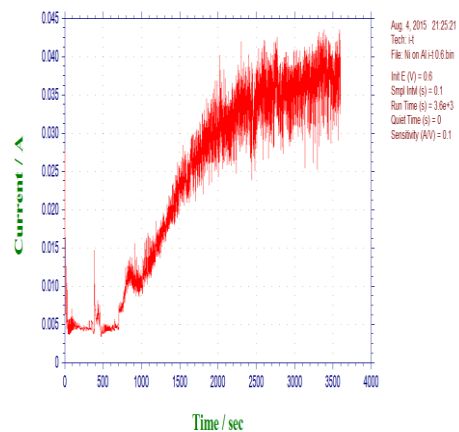
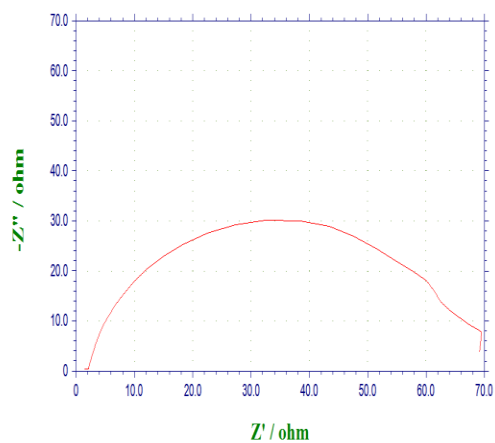
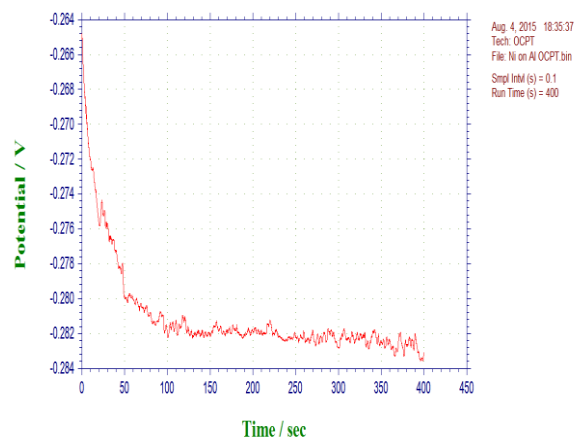
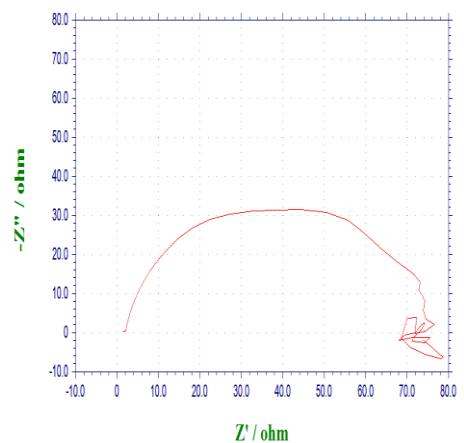
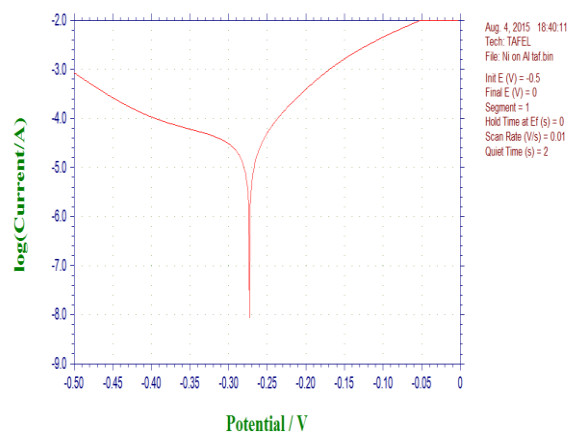
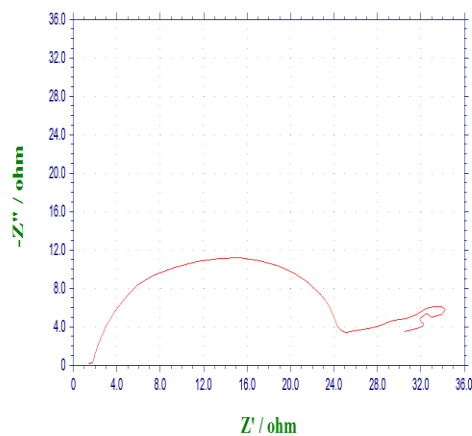
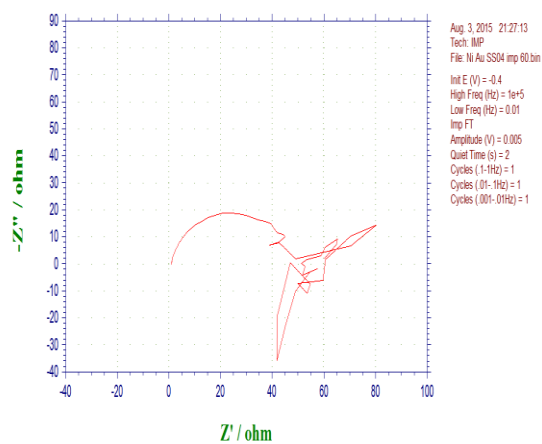
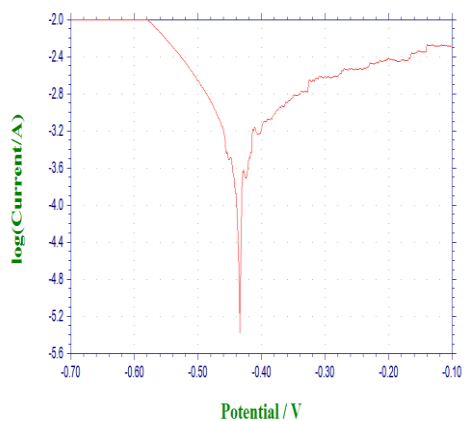
verified a drastic decrease of the corrosion current density. On the other hand, under potentiostatic conditions there was a three-fold increase in the current density at the cathode simulating operation. This behaviour was related by the authors to pitting the corrosion due to the penetration of the electrolyte through Plating defects. The same authors [59] evaluated a TiN-coated martensitic stainless steel bipolar plate and found that the corrosion rate was decreased by two orders of magnitude in comparison with the uncoated material. Jeon et al. [60] Investigated the influence of the  $N_2$  gas pressure during the deposition process over the corrosion resistance of TiN-Plated 316L stainless steel under simulating PEM fuel cell environment. They found marked differences in the corrosion current density and charge transfer resistance values depending on the  $N_2$  pressure. The variations were ascribed to the porosity of the coatings which were dependent on the  $N_2$  pressure employed during the deposition process. The best performance was related to the coating with the lowest porosity percentage. This result was confirmed through SEM images of the Platings surfaces.

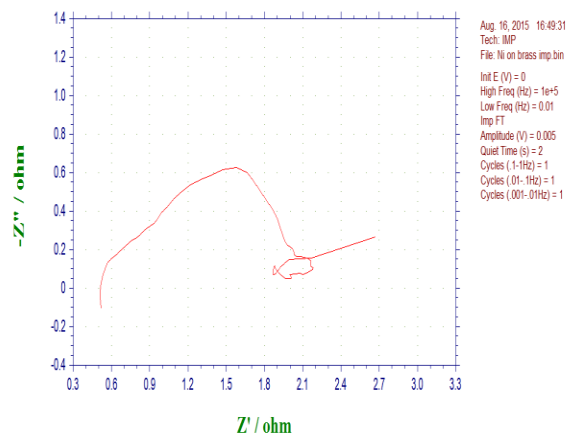
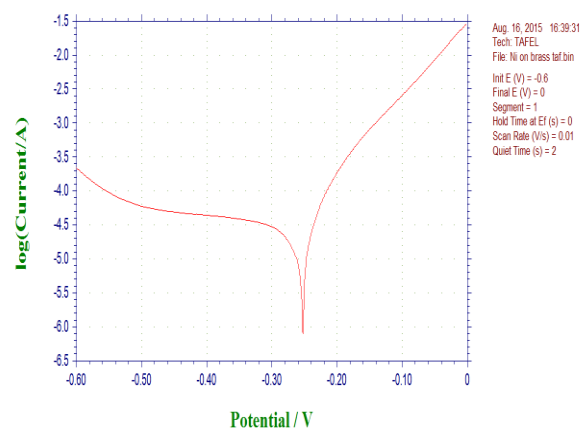
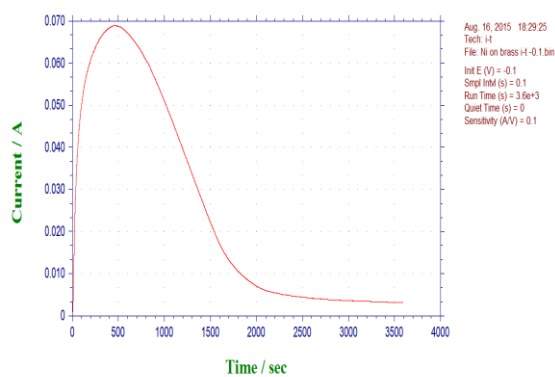
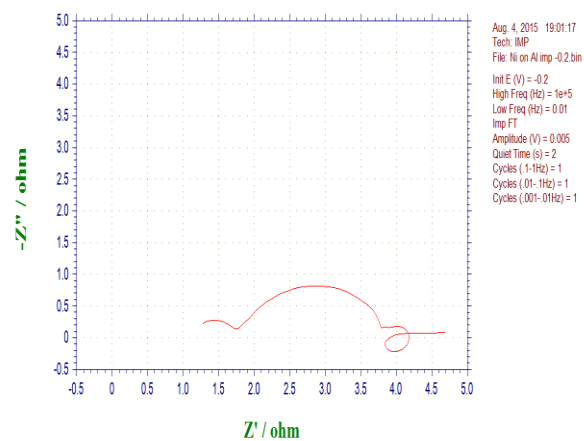
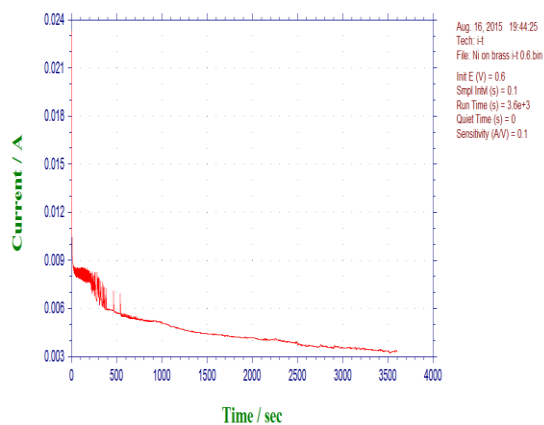
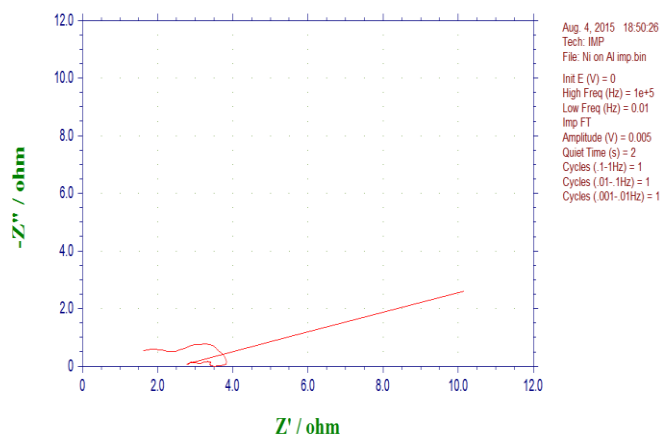
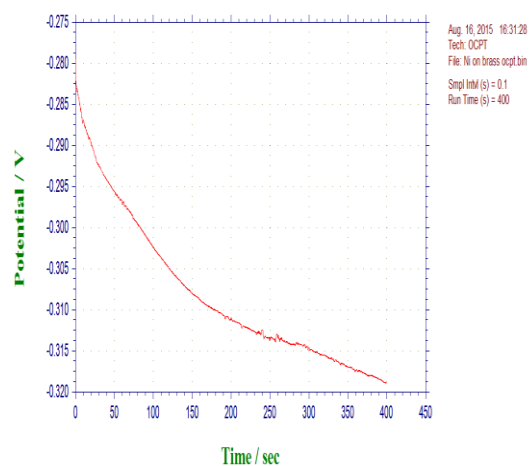
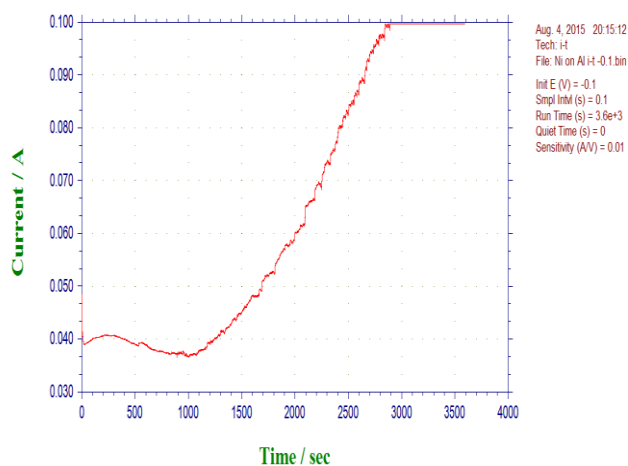
Different low cost metals are selected and the precious and non-precious metal platings has are done and it's corrosion resistant is evaluated in order to use the same as alternate Bipolar Plates in the PEM Fuel Cells.

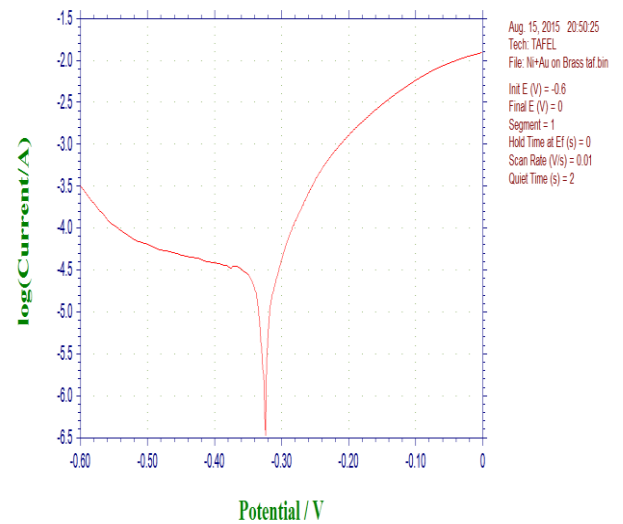
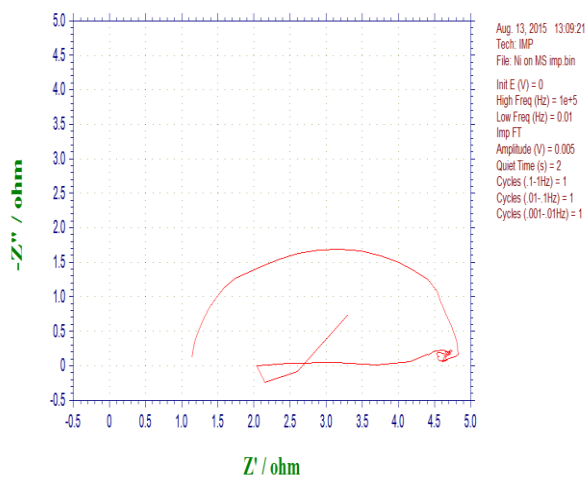
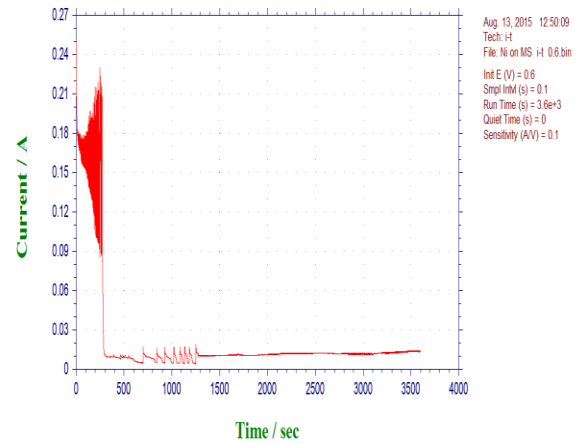
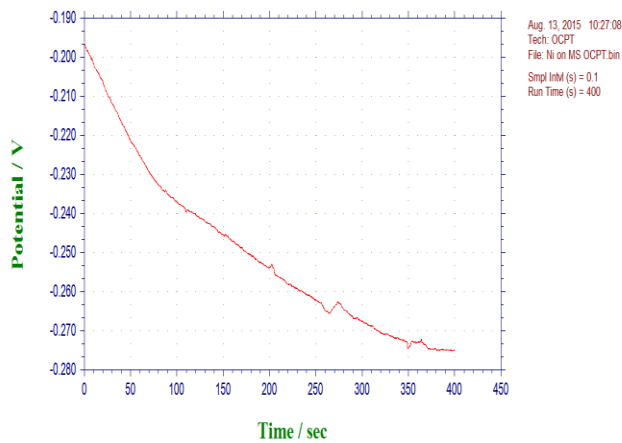
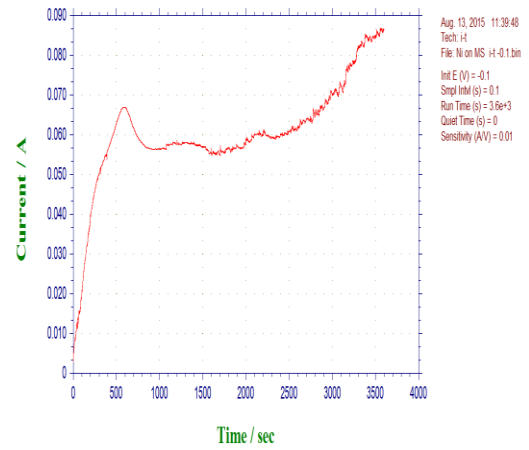
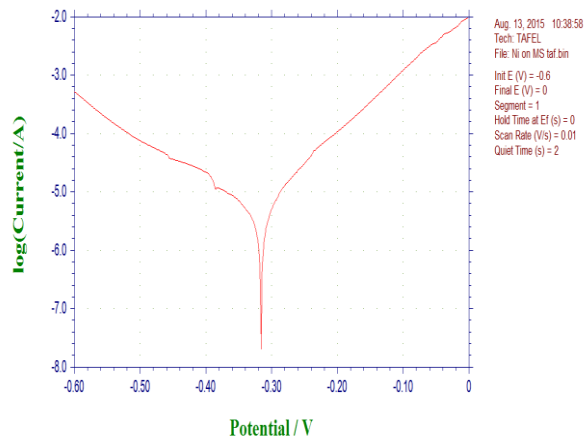
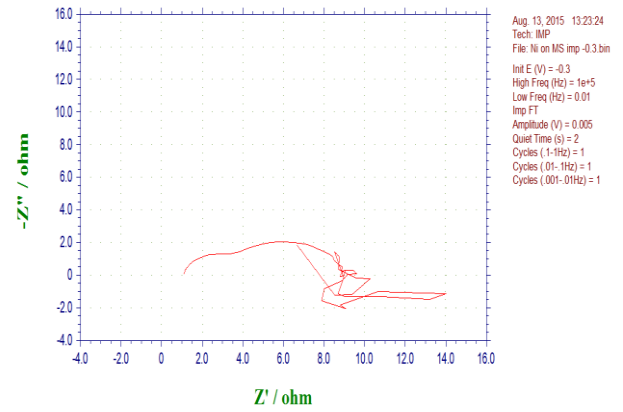
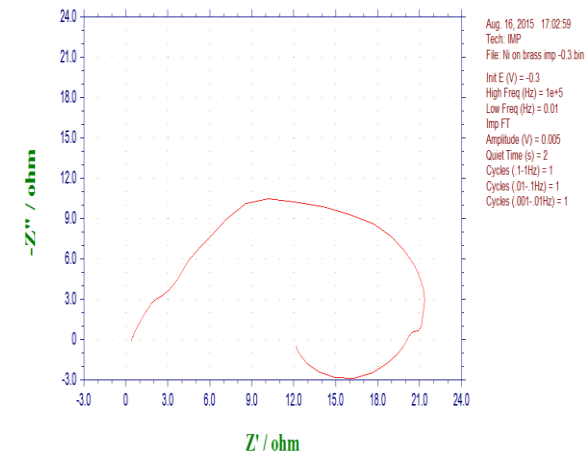
Sl.No	Plating Method	Plating Processes followed
1	Gold Top Coat Layering	Pulse Current Electrode Deposition
2	Stainless Steel Layering	Physical Vapor Deposition (PVD) (Example, Magnetron Sputtering) or Chemical Vapor Deposition (CVD) and Electro less Depositions for Ni-Pt Alloy.
3	Titanium Nitrate Layering	RF-Diode Sputtering
4	Nickel Layering on Aluminium Substrate	Chemical Vapor Deposition (CVD)
5	Nickel Layering on MS	Chemical Vapor Deposition (CVD)
6	Nickel Layering on Brass	Chemical Vapor Deposition (CVD)
7	(Nickel+Gold) Layering on Brass	Physical Vapor Deposition (PVD) (Example, Magnetron Sputtering)
8	(Nickel+Gold) Layering on MS	Physical Vapor Deposition (PVD) (Example, Magnetron Sputtering)
9	(Zinc+Gold) Layering on Aluminium	Physical Vapor Deposition (PVD) (Example, Magnetron Sputtering)
10	(Nickel+Gold) Layering on Galvanized Iron	Physical Vapor Deposition (PVD) (Example, Magnetron Sputtering)
11	(Nickel+Gold) Layering on SS304	Physical Vapor Deposition (PVD) (Example, Magnetron Sputtering)



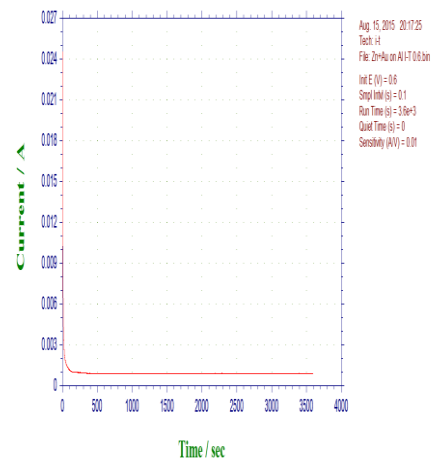
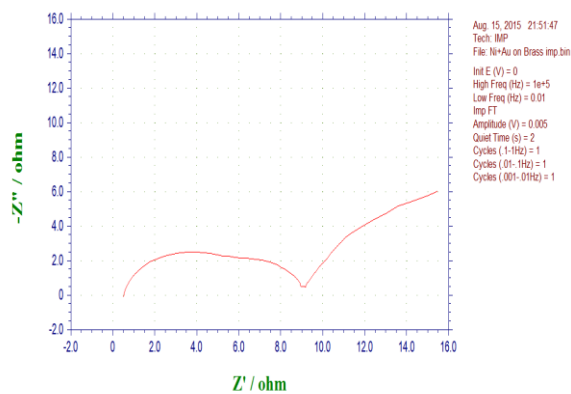
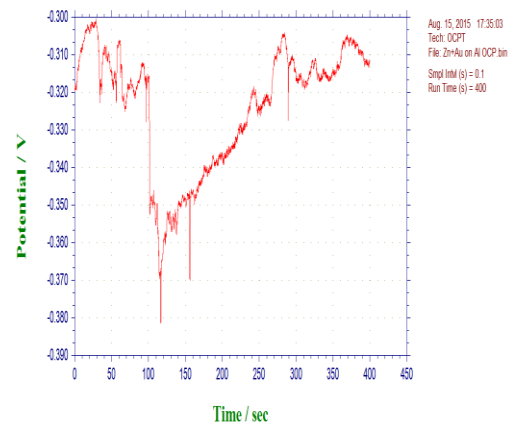
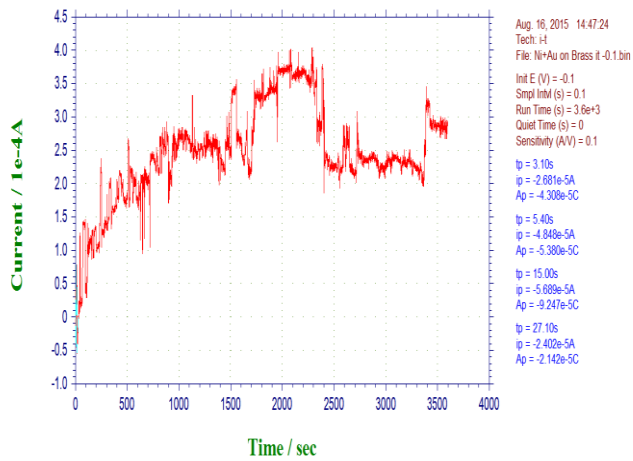
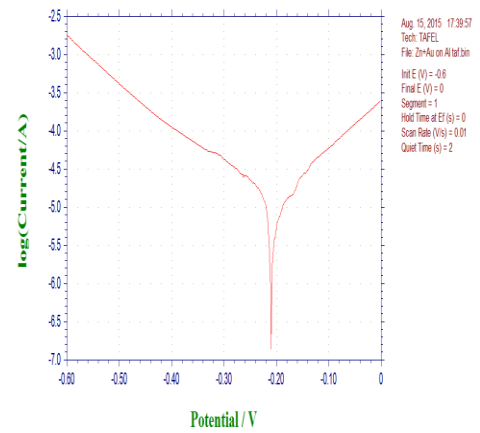
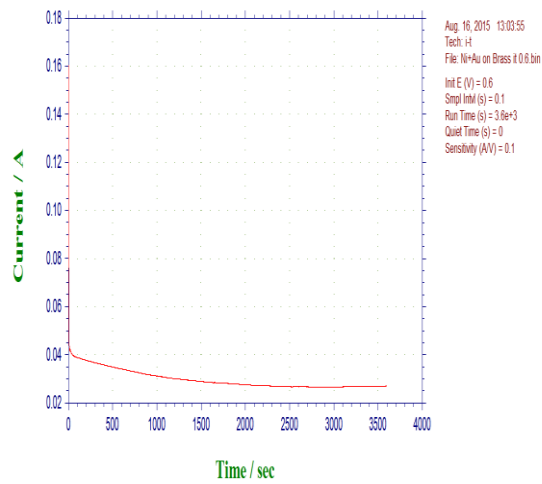
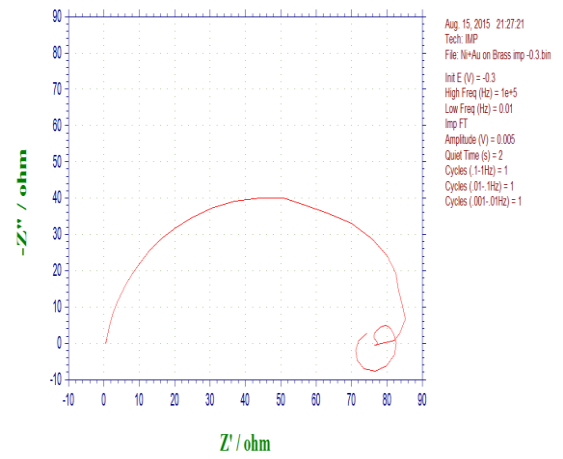
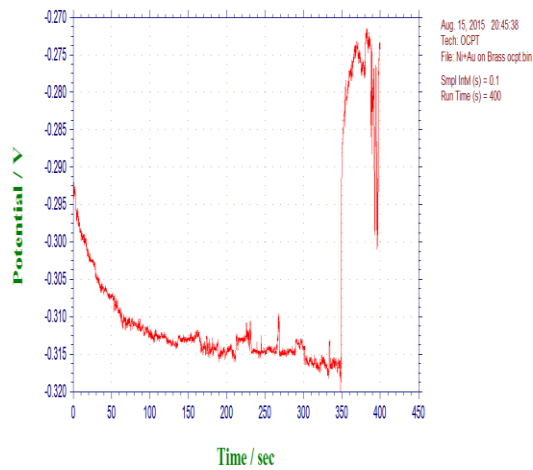


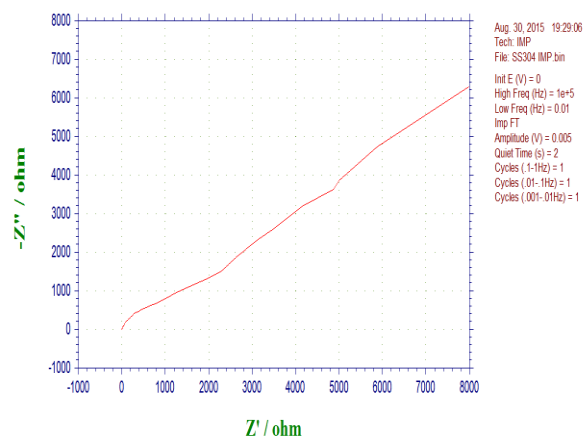
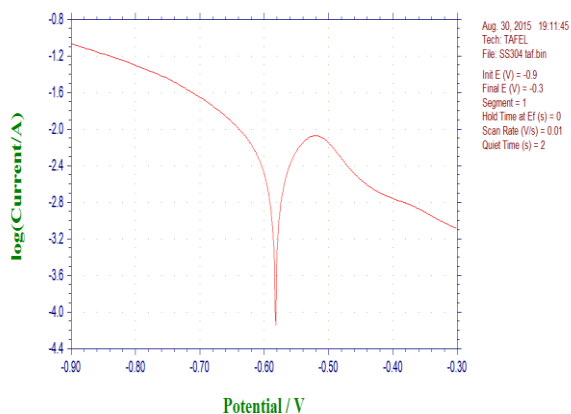
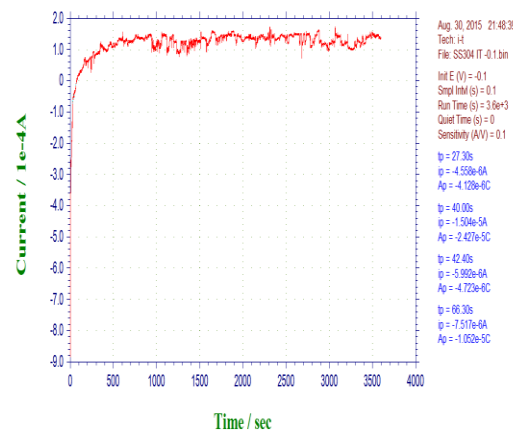
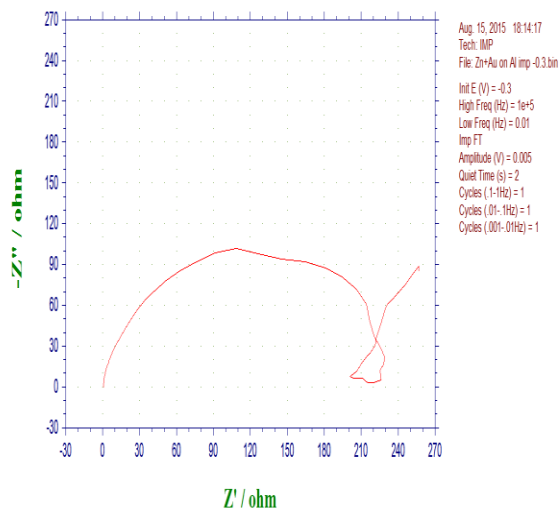
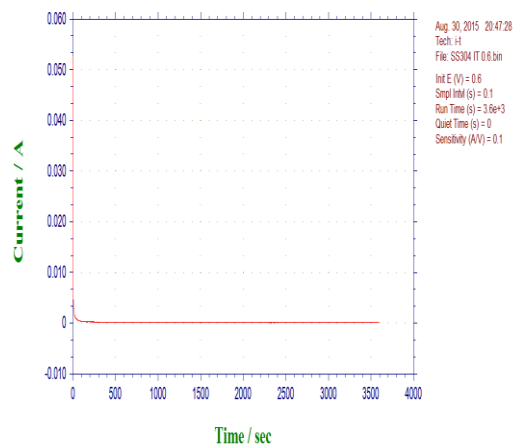
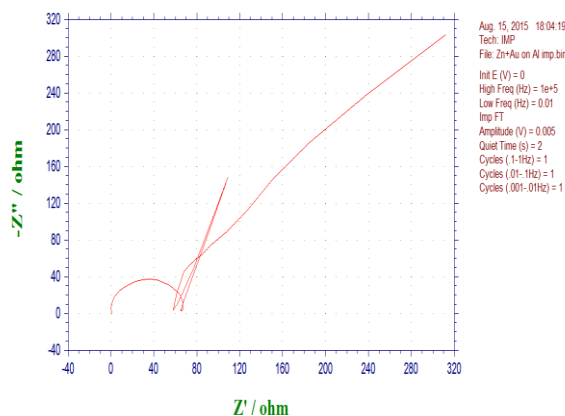
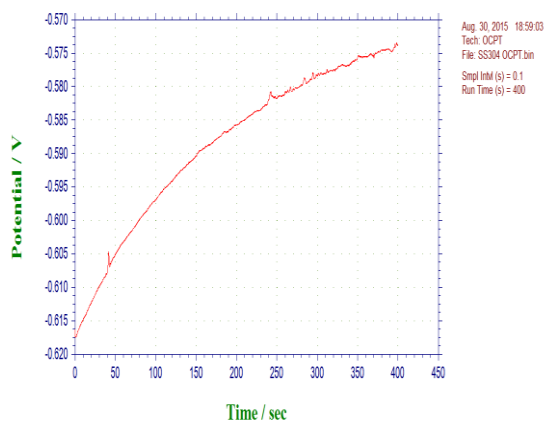
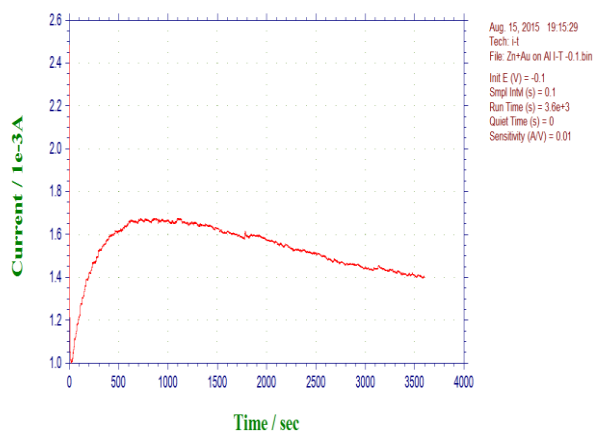


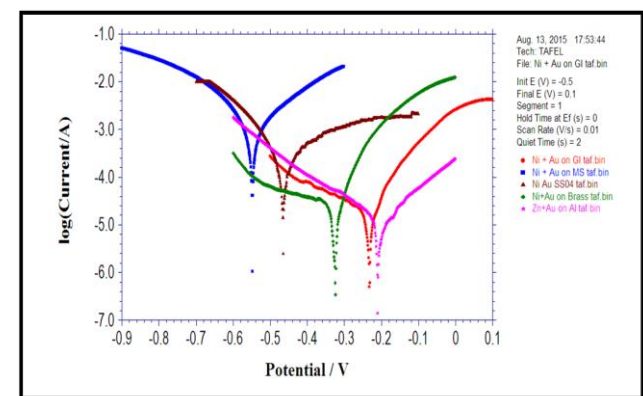
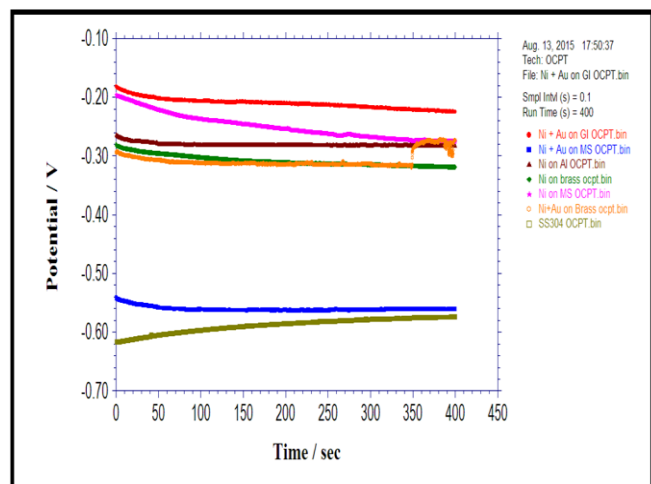
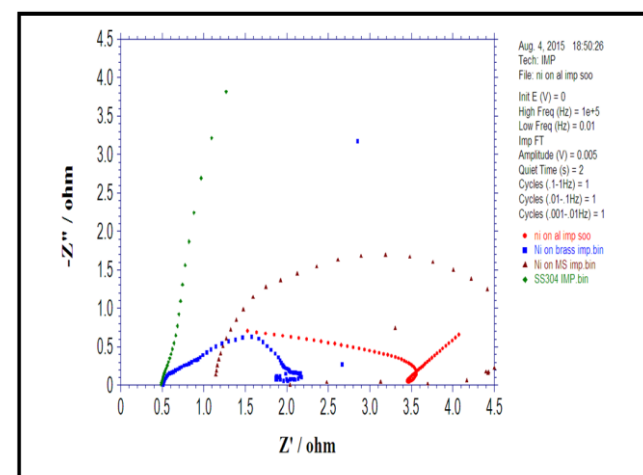
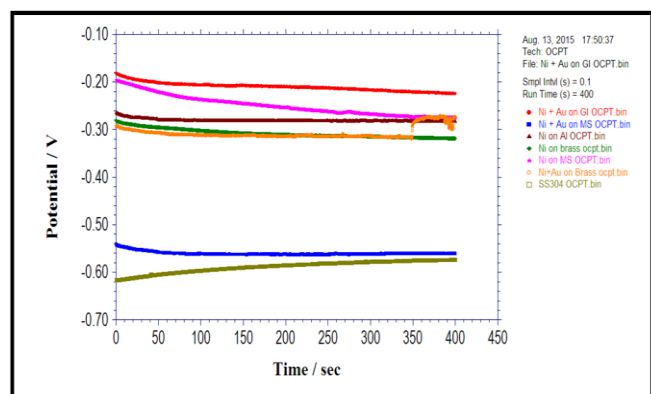
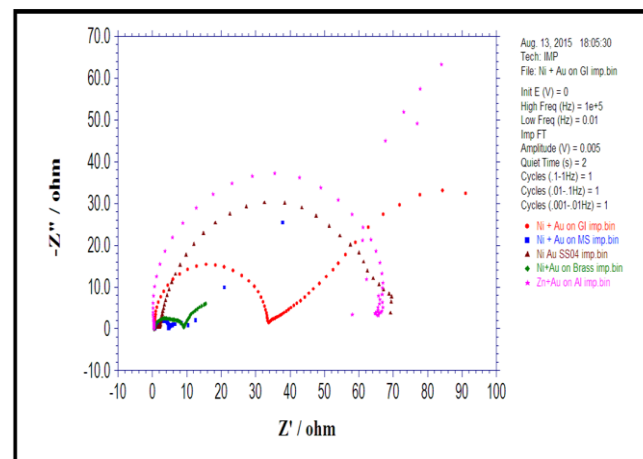
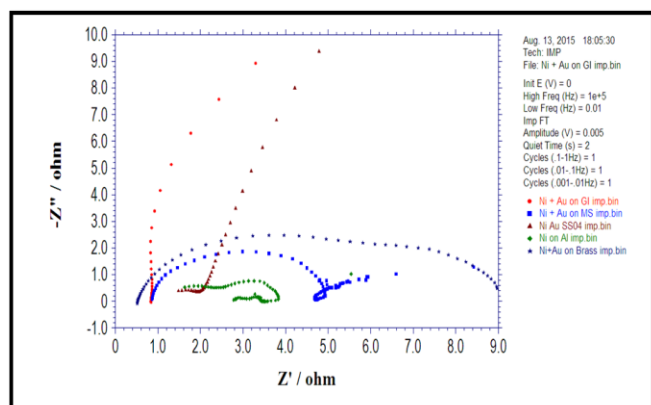
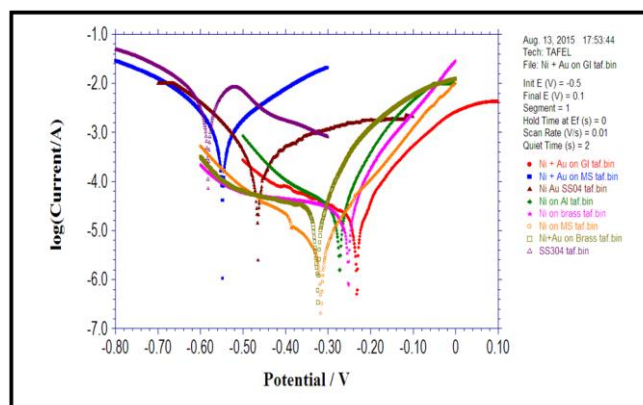
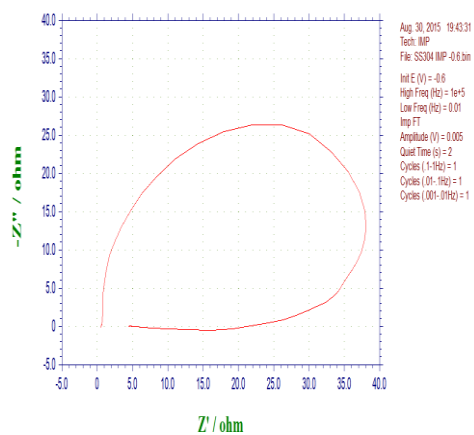


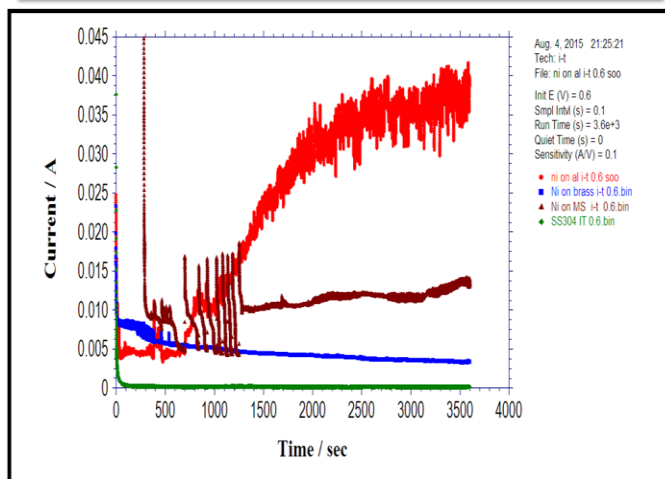
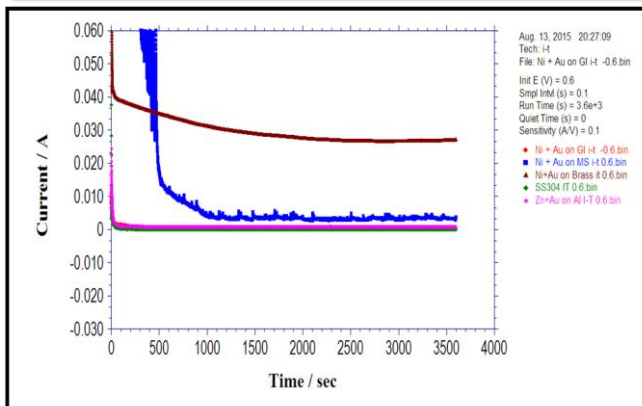
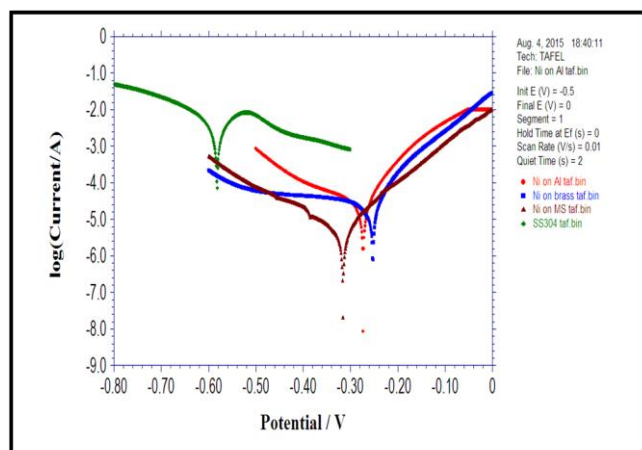








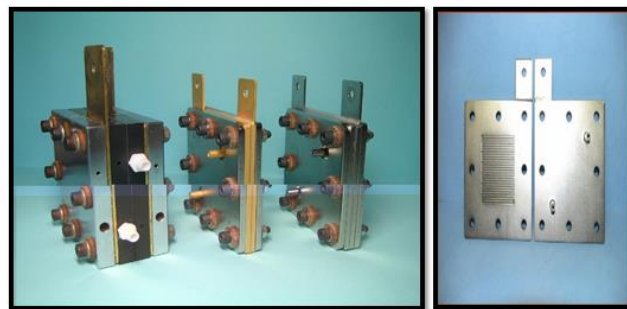




It is extensively documented that the presence of inherent defects on PVD coatings is a major concern as the corrosion resistance of the metal substrate is adversely affected by these imperfections [61–63]. In order to overcome these drawbacks other deposition processes have been tested on the bipolar plate stainless steels. Myung et al. [64] applied TiN nano particles on the 310S stainless steel via an electrophoretic deposition (EPD) process. Field emission SEM images showed that the nano-particles were still covering the stainless steel surface after 300 h of a normal PEM fuel cell operation. As a consequence of this relative stability the corrosion current density and contact resistance of the coated material were low, similarly to the pure graphite bipolar plates. Multi-layered PVD coatings are often reported as a way of achieving defect-free, corrosion resistant coated metal plates [65,66]. Based on these findings Ho et al. [67] performed corrosion tests with Ti/TiN and Ti/CrN Plated 304 stainless steel bipolar plates.

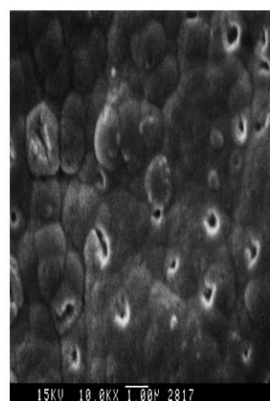
### Gold Plating – Process Specifications

Bias Power Supply	10 KW
Pulsed DC - Advance Energy	
Frequency	150 KHz
Duty Cycle time	0.5 Micro Secs
Voltage	350 to 600 Volts
Etch Current	6.5 to 9.0 Amps
Etching Duration	12 Mins
Ramping Duration	4 Mins
Argon Gas flow	300 SCCM
Glow Vacuum	2.2 Pa

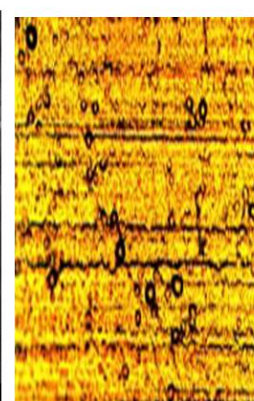


Prototype one-cell PEM fuel cell stack with Graphite bipolar/end plates (left hand side), Aluminum Bipolar/end plates coated with Gold (Au) (middle side) and Pure Aluminum Bi-Polar/end plates (right hand side).

### SCANNING ELECTRON MICROSCOPE IMAGES OF Bi-POLAR PLATES



SEM image of Pure  
Aluminum Bipolar plates



Photomicrographs  
of Gold-Plated

### Scale up issues and thermal management:

PEMFCs unlike other types of the fuel cells show some loss of efficiency and power density with the scale up of areas of electrodes and increase in number of cells in a stack. The main reason is that remove of the product-liquid water-becomes difficult in large systems; further, a high water vapor pressure in the reactant flows, causes an increase in over-potential, especially at the cathode.

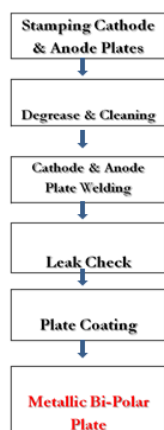
In addition, water condensation, electrode flooding and occlusion of the gas channels occur, leading to the operational failure. An issue strongly related to the water management is that of thermal management in stacks; on one hand, too low temperatures cause water condensation problems, but on the



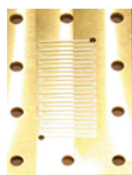
other hand, even more important, is that high cell temperature (even in confined areas) leads to membrane dehydration and consequent loss of performance. The 'rule of thumb' for the evaluation of heat losses in PEMFCs is that the power dissipated as heat is approximately equal to the electrical power supplied to the external circuit (deviations from this simple rule is noticed at high current densities and low voltages).

### Various Stages/Steps in Making/Developing the Required Bi-Polar Plates for Research Work

#### METALLIC BI-POLAR PLATE



#### GOLD PLATED ALUMINIUM



Pure Aluminium Bi-Polar/end plate with machined multi-parallel channel gas flow-field design designed and developed.

#### PEM FUEL CELL – MAJOR COMPONENTS

- ✓ Electrode – Pt catalyst used
- ✓ Membrane – “NAFION”  
Most commonly used
- ✓ Bipolar Plate – Graphite
- ✓ Membrane and Electrode



Gold Plated Aluminum



Gold Plated Aluminum Bipolar Plates Shown in Gas Manifold



GOLD PLATED ALUMINIUM BI-POLAR PLATE

#### DEVELOPED GOLD PLATED ALUMINUM BI-POLAR PLATE



Gold-Plated Aluminum Bipolar/end plate with machined multi-parallel channel gas flow-field design designed and

#### PURE ALUMINIUM BI-

#### GOLD PLATED ALUMINIUM



PROTO TYPE ONE CELL PEM FUEL CELL STACK WITH GOLD PLATED ALUMINIUM



#### Design criteria for bi-polar plate materials.

Sl.No	Material Selection Criteria	Limit
1	Chemical Compatibility	Anode face must not produce disruptive hydride layer. Cathode face must not passivate and become non conductive.
2	Corrosion	Corrosion rate $< 0.016 \text{ mA cm}^{-2}$
3	Cost	Material + Fabrication, US\$ $0.0045 \text{ cm}^{-2}$
4	Density	$< 5 \text{ g cm}^{-3}$
5	Dissolution	Minimization of Dissolution (For Metallic Plates)
6	Electronic Conductivity	Plate Resistance $< 0.01 \text{ ohm cm}^{-3}$
7	Gas diffusivity/ impermeability	Maximum average gas permeability $< 1.0 \times 10^{-4} \text{ cm}^{-2}$
8	Manufacturability	Must be low with high yield
9	Recyclable	Material can be recycled during vehicle service, following a vehicle accident or when vehicle is retired.
10	Recycled	Made from recycled material
11	Stack Volume/kW	Volume $< 11 \text{ kW}$
12	Strength	Compressive Strength $22 \text{ lb-in}^{-2}$
13	Surface Finish	$> 50 \text{ micro meter}$
14	Thermal Conductivity	Material should be able to remove heat effectively.
15	Tolerance	$> 0.05 \text{ mm}$



**Corrosion Data for Electrolytic Platings on Aluminium**

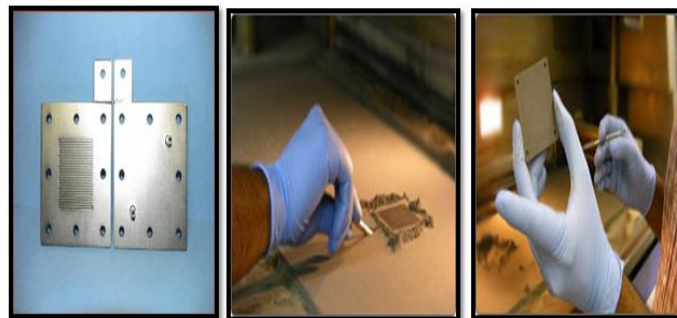
Sample	Corrosion Rate ( $\mu\text{m/year}$ )
4a. Gold over aluminium, as plated	~ 3000
4b. Gold over aluminium, thermally-cycled	~ 2000
4c. Gold over nickel over copper over aluminium, as plated	~ 1000
4d. Gold over nickel over copper over aluminium, thermally cycled	~ 750

The Platings have been deposited using a cathodic arc deposition system. The good corrosion resistance of the multi-layered coatings has been confirmed as the corrosion current densities were lower for the material coated with these coatings in comparison with the bare or single layer coated one. This behavior is described to the structure of the multi-layered Platings that inhibit the direct path between the corrosive environment and the steel substrate. Choi et al. [68] showed that the ternary (Ti,Cr)Nx Platings produced by the Inductively Coupled Plasma Magnetron Sputtering enhance the corrosion and electrical performance of 316L stainless steel in a simulated PEM fuel cell environment. The coating prevents the formation of oxygen on the surface of the material and act as an effective barrier between the substrate and the electrolyte. Fukutsuka et al. [69] used plasma assisted chemical vapor deposition process to produce a carbon layer on 304 stainless steel bipolar plate. The corrosion rate of the coated material was less than  $1\text{ mA cm}^{-2}$  reaching therefore the US DOE technical target for the bipolar plates [70]. Chung et al. [71] also evaluated the corrosion performance of a carbon coated 304 stainless steel bipolar plate. The carbon layer was produced via a chemical vapor deposition method using a  $\text{C}_2\text{H}_2/\text{H}_2$  mixed gas as carbon source. Depending on the acetylene to hydrogen ratio the coating morphology varied from a filamentous porous layer to a continuous carbon coating. For the latter morphology the corrosion resistance was found to be high and reaching the performance of commercial pure graphite bipolar plate (Poco graphite). Fu et al. [72] found that a C–Cr composite coating greatly decreased the interfacial contact resistance and corrosion rate of 316L stainless steel. They used a pulsed bias arc ion plating (PBDIP) deposition method which was chosen due to the low temperature, dense layer with few droplets that favour the final corrosion performance of the base metal. Feng et al. [73] investigated the performance of an amorphous carbon coated 316L stainless steel bipolar plate in a PEM fuel cell environment. They reported promising results of corrosion resistance under potentiodynamic and potentiostatic conditions and interfacial contact resistance. The dense and compact nature of the deposited film allied with the intrinsic high chemical stability and electrical conductivity of the carbon layer are responsible for the high desirable performance of the amorphous carbon coating.

**Aluminium**

Aluminium may be considered for bipolar plate applications in PEM fuel cells due to the low manufacturing cost [89]. It is well propelled, though, aluminum and its alloys corrode in the fuel cell environment leaching ions that contaminate the membrane, decreasing the cell output [90,91]. Regardless this limitation, coated-aluminium bipolar plates may reach DOE technical targets [92]. Joseph et al. [93] found that PANI-coated 6061 Al alloy showed very good

corrosion resistance compared to the non-plated substrate. The corrosion rate was up to  $10^{-8}\text{ A cm}^{-2}$  for a 40mm thick layer.

**PEM FUEL CELL – MAJOR COMPONENTS**

- ✓ Electrode –Pt catalyst used
- ✓ Membrane– “NAFION” Most commonly used
- ✓ Bipolar Plate–Graphite

**Gold Plated Aluminum****Gold Plated Aluminum Bipolar Plates Shown in Gas**

However, no information about coating degradation in long-term immersion tests is reported in the paper. PPy coating, on the other hand, presented a corrosion current density of the same order of magnitude than that of the bare alloy. According to the authors the lack of corrosion resistance in the case was due to the pinholes on the polymer layer after deposition.

**Effects of ion contamination on PEM fuel cell performance**

Notwithstanding the crucial role of the corrosion processes over the increase of the interfacial contact resistance of metal bipolar plates the effects of the resultant cationic contaminants on the membrane conductivity should not be neglected. It is very probable that, in real PEM fuel cell systems, contaminants originated from several different sources poison the membrane decreasing the overall power output. Cheng et al. outlined all these sources of membrane contamination in a recent review paper [107]. Cationic impurities are released after corrosion of bipolar or end plates and even of the fuel cell piping system. Collier et al. [108] cited the presence of  $\text{Fe}^{3+}$ ,  $\text{Cu}^{2+}$  and other cations in a unit cell MEA after 10,000 h of operation. According to Cheng et al. [107] almost all cations exhibit higher affinity for the sulfonic groups in the polymeric membrane than  $\text{H}^+$ . Hence, when other cations exchange for the protons in the polymer structure, the amount of water in the cell is reduced

**Conclusions**

There is a wide variety of materials and surface modification methods available for the metal bipolar plate development. It is evident that the major challenges regarding to a successful commercial use in PEM fuel cells are related to the increasing corrosion resistance and decreasing contact resistance at a reasonable cost. The Table 2 summarizes the literature findings on the corrosion data found on all the papers reported in the review. Few materials present a suitable combination of corrosion current density and contact

resistance according to DOE targets. Depending on the reference consulted different values of corrosion current density have been found for the same material, especially for 316LSS. These values may vary as a function of electrolyte composition, pH and temperature, specimen surface finishing and small variations on the composition of the material itself. Although there is a plenty of information on the corrosion resistance of the metal bipolar plates, long term results are often missing on the most part of the literature reports.

### Sustainable Energy Development

The quality of life is dependent on access to a bountiful supply of cheap energy. For a sustainable future, energy should be derived from non-fossil sources; ideally, it should also be reliable, safe, flexible in use, affordable and limitless. Energy resources have always played an important role in the development of the human society. Technological developments and consumption of energy and increase of the world population are interdependent.

It is known fact that,

- ✓ 8 countries have 81% of the global crude oil reserves;
- ✓ 6 countries have 70% of all natural gas reserves and
- ✓ 8 countries have 89% of all coal reserves.

It must be considered that even if the bipolar plate presents the corrosion current density slightly higher than the DOE target of 1 mA cm<sup>-2</sup> it is not necessarily true that its behavior will not be acceptable during the long term operation. The overall performance must take into account the fuel cell output which, in turn, depends on the interfacial contact resistance and contamination of the membrane with metal ions produced by the corrosion processes. The search for coatings or surface modification treatments that are capable of increasing bipolar plate corrosion resistance and simultaneously decrease contact resistance is a very well established trend. Interesting results have been reported on nitriding, PVD or CVD Platings and passive film modification. The main challenges are to produce defect-free Platings, stable passive films or nitride layers that are able to protect the metallic substrate from the fuel cell harsh environment.

Different low cost metals has been selected and the precious and non-precious metal platings has been done and it's corrosion resistant is evaluated in order to use the same as an alternate Bipolar Plate in the PEM Fuel Cells. Among all the low cost metals, Aluminum playing a vital role and Gold Plated Aluminium Substrate provides higher corrosion resistance and the same can be used as a Metallic Bi-Polar Plate for PEM Fuel Cells.

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