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INVESTIGATION MANUFACTURE AND PROPERTIES OF INCONEL ALLOY PLATE WITH CONDUCTIVE COATING USED FOR METALLIC BIPOLAR PLATES MEMBRANE FUEL CELL

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Abstract

One of the components of these cells is bipolar plates. These plates have a considerable effect on the weight, volume and the cost of these cells. According to the materials that used in their manufacturing, the bipolar plates categorize to: metallic, composite and graphite plates. One of the disadvantages of composite and graphite plates is the higher weight and volume of cell's stack because of small deformation ability and weak stampability property of these plates. So the industrial use of metallic plates is usual. The disadvantages of metallic plates are Low corrosion resistance in acidic environment, increasing of the interfacial contact resistance caused by presenting of the oxide layers on the surface of metallic plates and creating of ionic contamination caused by corrosion products. In this study, the Inconel 625 is selected because of high corrosion resistance of this metal. Interfacial contact resistance of the Inconel 625 is investigated in different temperatures. The results show that by increasing of temperature and time, the amount of interfacial contact resistance is increased. According to this test, the TiN

coatings that is created by PVD (cathodic arc evaporation), cause that the corrosion resistance and interfacial contact resistance of samples are improved. X-Ray Fluorescence, X-Ray diffraction, Energy dispersive X-ray spectroscopy, Scanning electron microscopy and contact resistance and corrosion potentiodynamic polarization tests are used for investigating of samples. Totally, results show that the amount of the corrosion resistance and interfacial contact resistance of coated samples, are lower that amount that are reported by energy department of America. By the way, the corrosion resistance and interfacial contact resistance of sample with 1200 nm respectively. This sample and 8.14 $m\Omega.cm^2$ coating thickness and Ra= 400, are 0.1843 $\mu A.cm^{-2}$
Key words: Fuel cell, bipolar plate, inconel has the most suitable situation for using in fuel cell 625, titanium nitride, cathodic arc, contact resistance evaporation, corrosion resistanc

1. Introduction

Bipolar plates in polymer membrane fuel cells (PEMFCs), are responsible for various tasks. Such responsibilities includes the distribution of fuel and oxidant in the mass, transporting reaction products, conduction electrons and etc. Thus, selecting the type of bipolar plates with high ability of functioning in fuel cell conditions is very important. Traditionally, PEMFCs are made of graphite or graphite composites with excellent corrosion properties, chemical stability, high conductivity and low contact resistance. But, these materials are expensive and have poor mechanical properties. This issue limits further applications of graphite as a material for the page. So, investigation to solve such problems is imperative [1-3].

In recent years, the application of PEMFCs are more developed and has attracted more attention. Recently, metallic bipolar plates as the material for bipolar plates are developed due to the high mechanical strength, low gas permeability, very high thermal and electrical conductivity, the ability of being marked and relatively low production costs [1-4]. Because, fuel cells indoor temperature is about 80 °C and the PH is between 2 to 3 [5], Corrosion of the metal plates in fuel cells is very high and there are many problems to come (serious hindrance) that are effective in the performance and durability of fuel cells. This issue leads to the formation of a passive layer on the surface of the bipolar plates and as a result, high contact resistance would be expected which reduce the conductivity of bipolar plates and fuel cell performance. Also, the corrosion products (mainly various Cations), infect the membrane and catalyst and affect them on routine tasks because the polymer membrane is a strong Cation exchanger and the catalyst is exposed to ion contamination. Therefore, based on these two important issues (High corrosion resistance and low contact resistance), adding a corrosion resistant coatings on metal plates surely improve long-term performance and durability of a mass and this issue is inevitable [1, 6-8].

In recent years, nitriding the metals as a cover on the metal plates is highly developed [2]. The nitriding process is done through one of the two traditional heat treatment process or physical vapor deposition process (PVD), in order to cover a hard surface layer on metal components. Among the nitride coatings, the TiN coatings are more applicable due to their high corrosion resistance and electrical conductivity similar to metal [2, 8, 9]. This study investigates the corrosion resistance and interfacial contact resistance properties of TiN coatings on the Inconel 625 sub layer for using in bipolar plates in polymer membrane fuel cells.

2. Experiments

2.1. Materials

Table. 1 shows chemical composition of Inconel 625, which in this study is used as a substrate of the bipolar plates. The samples in size of 1 ×1 cm^2 were made of Inconel 625 sheets. In order to evaluate the effect of surface roughness of the substrate on the contact resistance, the samples were polished with different sandpapers (400, 600, and 1500). In order to determine the thickness of the contact resistance values and resistance to corrosion, Titanium Nitride coatings (TiN) in various thicknesses (400 nm, 800 nm, and 1200 nm), using Cathodic arc physical evaporation method were prepared on the surface of the samples.

Table 1
Chemical compositions of Inconel 625

Elements	Ni	Cr	Mo	Fe	AL	Nb	Ti
Percent (%)	66.5	25.5	4.2	5.2	0.081	2.8	0.413

2.2. Oxidation of uncoated samples

To study the effects of time and temperature on the surface oxidation of uncoated samples, and to assess the effects of oxidation on the conductivity of the samples, two experiments were designed. In the first experiment, the samples were placed inside the furnace at a constant temperature of 120 ℃ and variable time and then conductivity of the samples was measured. The conductivity of the samples was measured at the initial time. The samples were then placed into a furnace and after 12, 24, 48, 60, and 72 hours the conductivity was measured. Therefore, the effect of time on the surface oxidation and the oxidation effects on conductivity were checked.

In the second experiment, the time was stable and fixed but the temperature was changing. In this experiment, the sample conductivity was measured at 25 ℃. The samples were then placed into a furnace for 68 minutes and conductivity was measured at 80 ℃, 120 ℃, 200 ℃, and 300 ℃. Therefore, the effect of temperature on the surface oxidation was investigated.

2.3. Corrosion test

Corrosion properties of uncoated and coated samples, was examined by potentiodynamic polarization test. The conventional three-electrode system, includes platinum as a counter electrode (CE), saturated calomel electrode (SCE) as a reference electrode (Ref), and test sample as a working electrode (WE), was used for electrochemical tests. Corrosion cells, to measure the corrosion current density and corrosion of uncoated and coated samples, was connected to potentiostat devices, model of PGS-2065. In this experiment to simulate the acidic environment of the fuel cell, the 0.5 molar Sulfuric Acid + 2 ppm HF Acid were used as an Electrolytic corrosion test and the temperature of corrosion was 70 ±1℃. Corrosion tests in the potential range of -1 V to +1 V was done and the scanning velocity was 1mv.s^{-1}.

2.3.1. Interfacial contact resistance (ICR)

To measure the Interfacial contact resistance (ICR) of coated and uncoated samples of TiN, the way presented by Wang et al. [10] was used. Bipolar plate surface ICR of Inconel would be measured by putting the bipolar plate between the gas diffusion layers (GDL), under the real simulation condition and measuring the voltage drop ($R=VA_S/I$) between the samples. where R is the electrical contact resistance, V the voltage drop through the setup, I the current applied and A_S the surface area. In this experiment, the gas diffusion layer was used (Torey TGP-H-060). In this experiment the gas diffusion layer in contact with the two copperplates. The arrangements of surface contact resistance testing is declared in figure 1.

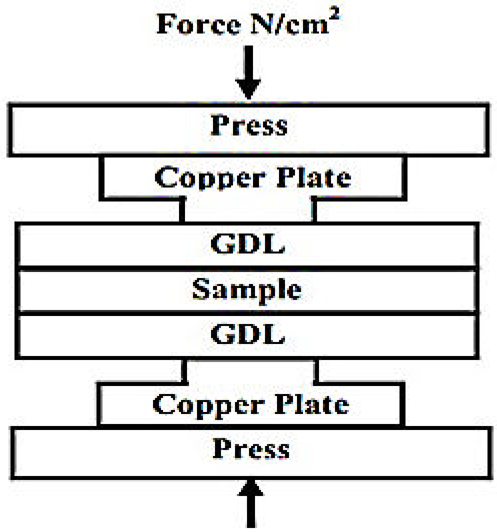


Fig. 1. Schematic of the test assembly for interfacial contact resistance

Applied compaction force during the experiment was at the range of 35 – 150 $N.cm^{-2}$. The overall strength of the arrangement, were recorded by Milliohm Meter device. The total measured of resistance (R_1), includes bulk resistance of the two copper plates ($2R_{Cu}$), two contact resistance between the GDL and copper plates ($2R_{GDL/Cu}$), two bulk Interfacial contact resistance of GDL ($2R_{GDL}$), two Interfacial contact resistance between the bipolar plates and GDL ($2R_{GDL/Bp}$), and the bulk resistance of bipolar plates (R_{BP}). The total resistance (R_1) is measured as follows: $R_1 = 2R_{Cu} + 2R_{GDL/Cu} + 2R_{GDL} + 2R_{GDL/BP} + R_{BP}$. As, the volume of resistance values of copper plates, carbon and Inconel samples is small compared with the existing contact resistance, they can be ignored. Therefore, the total resistance is equal to the sum of the Interfacial contact resistance. To calculate the Interfacial contact resistance of carbon plates with Inconel sample ($2R_{GDL/S}$), the Interfacial contact resistance of carbon plates with Copper plates ($2R_{GDL/Cu}$) must be measured. Do to so, the experiment of putting a carbon plate with a copper plate must be done again. The obtained resistance, is the resistance of Carbon plates with copper plates ($2R_{Cu/GDL}$). Therefore, ICR GDL with plate of Inconel with/out the coating is equal to $R_{BP/GDL} = (R_1 – R_2)/2$.

3. Results and Discussion

3.1. The results of X-ray diffraction

X-ray diffraction was carried out to determine the chemical composition of the coating. Results obtained from the analysis of titanium nitride coatings by the Cathodic arc evaporation method showed the existence of TiN on a substrate Inconel.

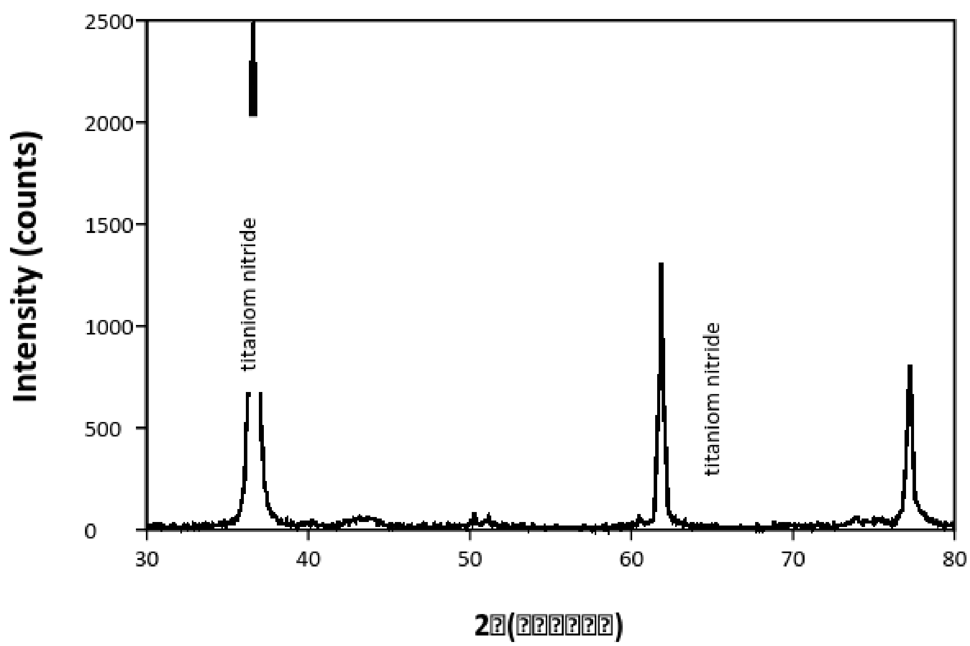


Figure 2. X-ray diffraction curves of TiN coatings on substrates Inconel.

3.2. Interfacial Contact resistance test results

3.2.1. Interfacial Contact resistance of uncoated samples

The Interfacial contact resistance of uncoated substrate of Inconel 625 is 17.87 $m\Omega.cm^2$. That is less than the amount set by the US Department of Energy (20 $m\Omega.cm^2$). The results of the Interfacial contact resistance for all uncoated samples show that the Interfacial contact resistance is created by increasing the applied pressure. This is because of the increasing of the contact pressure which makes GDL close to and the bipolar plates and the number of rough spots in contact with each other increase. And increasing the level of actual contact between GDL and the bipolar plates reduce the Interfacial contact resistance.

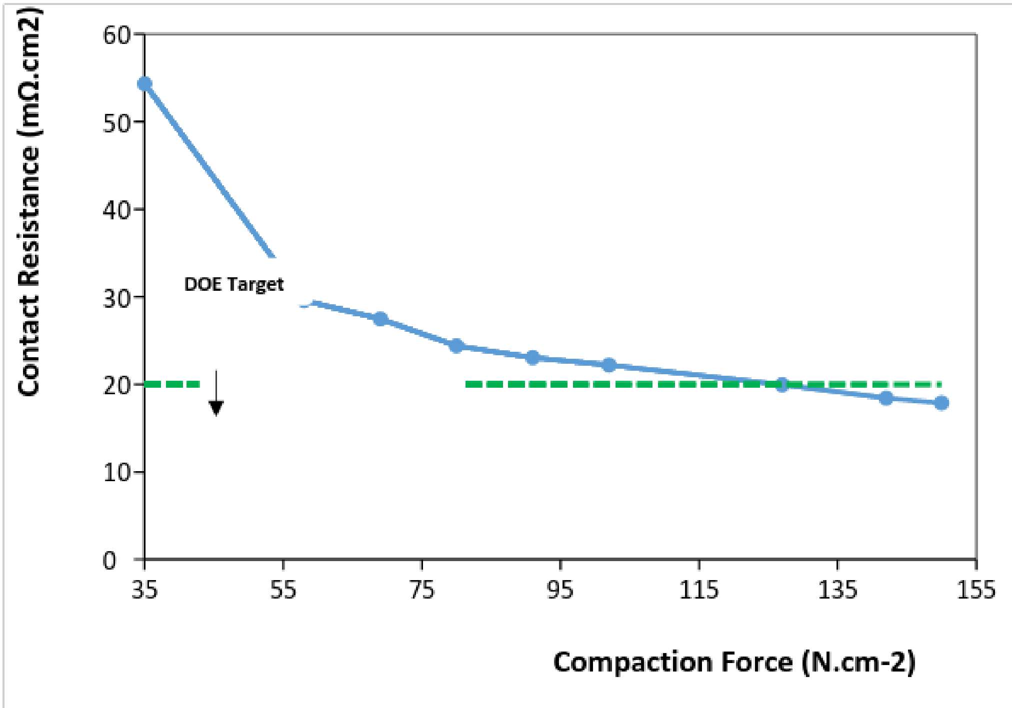


Figure 3. Uncoated Inconel alloy contact resistance.

3.2.2. Interfacial Contact resistance of oxidized samples in the furnace

The results indicated that the Interfacial contact resistance of the alloy of Inconel up to 80°C, was not increased and the amount was 26.9 mΩ.cm^2 and after that in the temperature of 200°C and 300°C, the amount was 124.42 and 606.19 mΩ.cm^2. This amount of Interfacial contact resistance, for the bipolar plates for PEM of fuel cells is very high. In another test, operating temperature of PEM fuel cell (120°C), was examined. The results showed that the Interfacial contact resistance of the alloy of Inconel at first was 31.89 mΩ.cm^2 and after the 72 hours, the amount was increased to 926.5 mΩ.cm^2. The results of these tests showed that the use of this alloy without coatings for using in bipolar plates is not possible. This is due to the formation of a passive oxide layer on the surface of the alloy of Inconel that increases the Interfacial contact resistance.

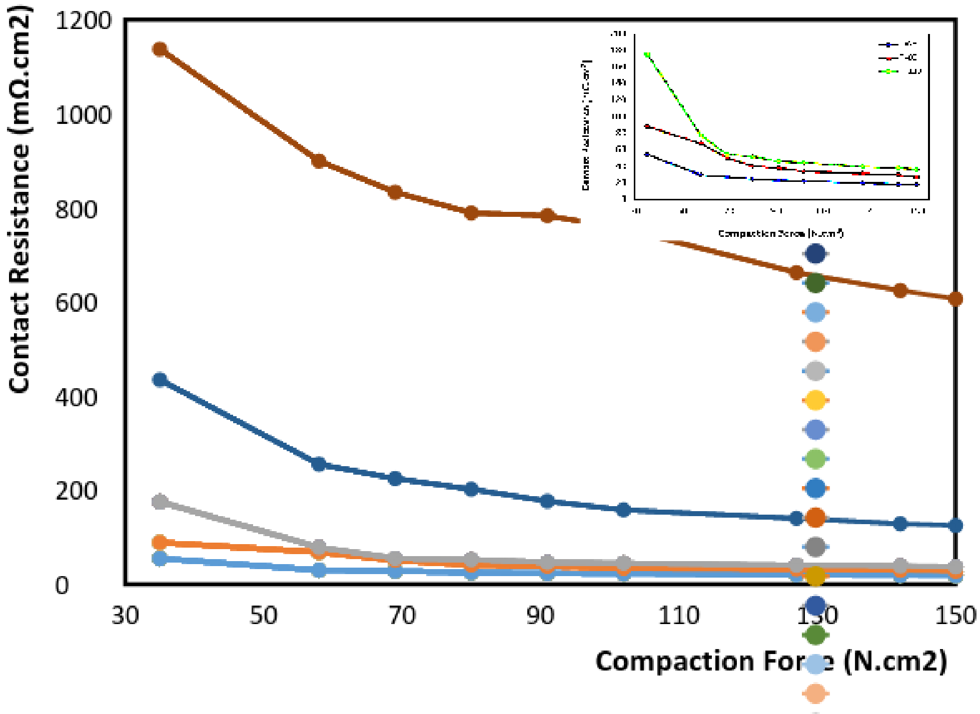


Figure 4. Effect of temperature on contact resistance of uncoated Inconel.

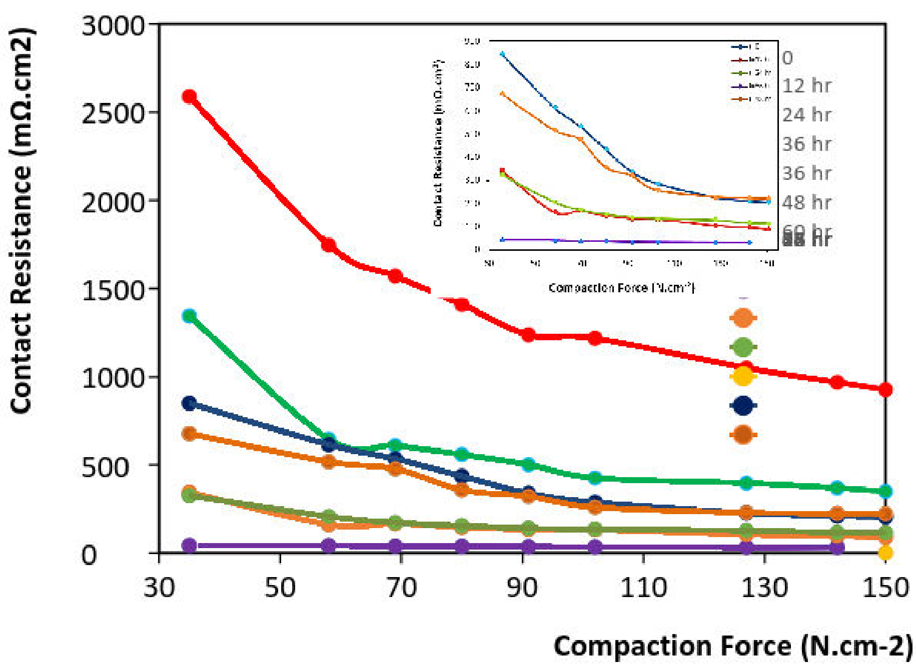


Figure 5. The contact resistance of uncoated Inconel ° C120 and at different times.

3.2.3. Interfacial Contact resistance of the TiN coating

The Interfacial contact resistance of coated substrates represents that contact resistance values was close to the values defined by US Department of Energy (DOE). The results indicate that using TiN coating on the alloy of Inconel improves Interfacial contact resistance. To evaluate the effect of coating thickness by evaporation arc cathode, subjects with a certain roughness were compared together the effect of coating thickness on these samples were analyzed. The amount of surface Interfacial contact resistance of the coating thickness was 400 nm, 9.3 $m\Omega.cm^2$ and then the thickness became 800 nm, 11.78 $m\Omega.cm^2$, and at the end the thickness became 1200 nm, and the amount was 10.12 $m\Omega.cm^2$. Evaluating the results of the effect of thickness on the surface layer of Inconel showed that at first by increasing the coating thickness to 800 nm, and after the increasing the thickness to 1200 nm, the Interfacial contact resistance decreased.

The results of the roughness of the substrate showed that the coating on the rough surfaces has less Interfacial contact resistance. The reason might be because of that the grosser levels, are important in coating. The rough surface of the substrate before coverage can increase the effective surface area of the substrate. It means that a constant cross section, contacts substrate with increased coating which can be increased to cover a greater in conductivity.

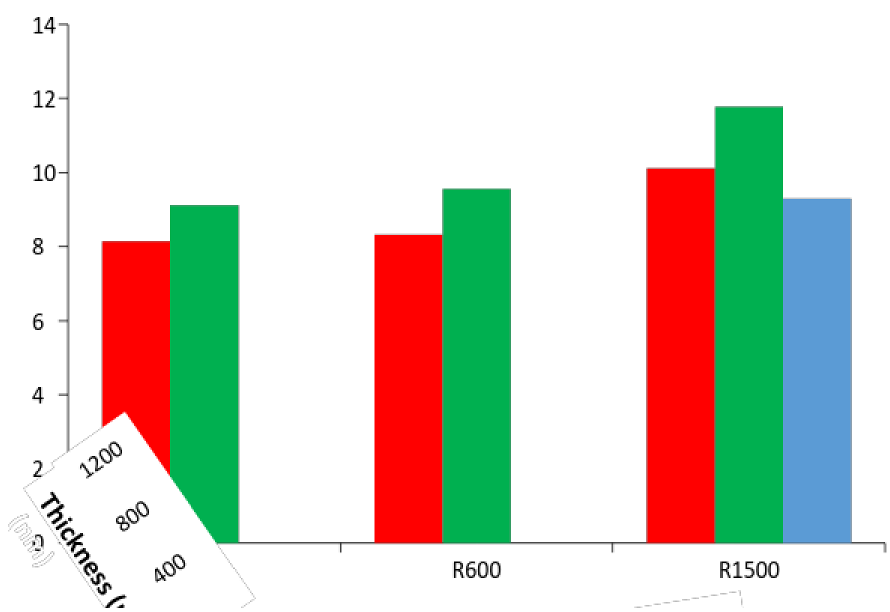
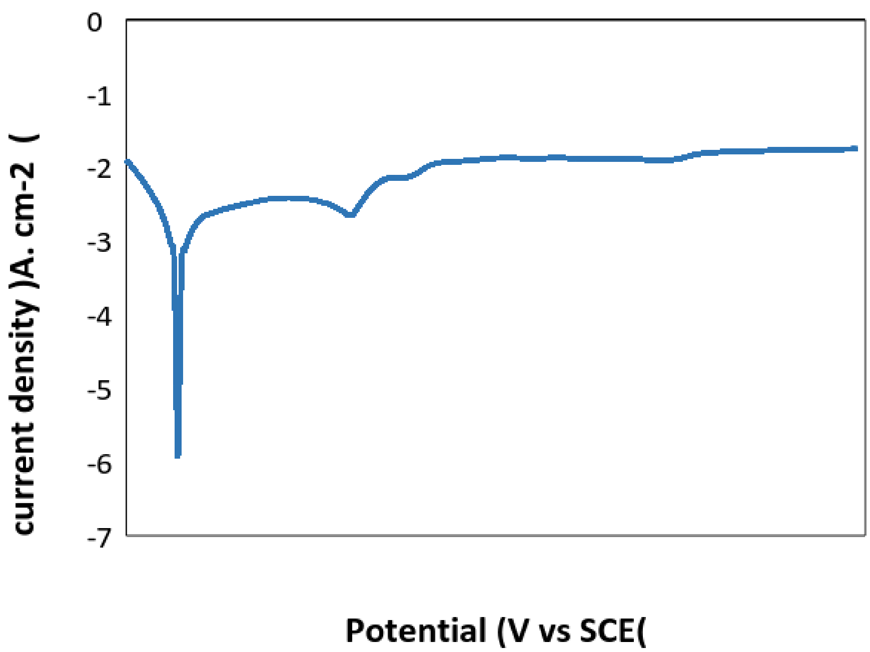


Figure 6. Effect of TiN coating thickness and surface roughness of the substrate on contact resistance Inconel alloy.

3.3. Corrosion test

3.3.1. The results of uncoated substrate corrosion

The results of the corrosion behavior of the alloy of the uncoated Inconel 625 show that a potential increase in the category of Andy make an increase in current and then by increasing the potential a lasting inactive state on the surface level. The passive layer on the surface, protect against the corrosive environment. According to the results of potentiodynamic polarization test, the corrosion current density for the Inconel 625, was 2.16 μAcm^{-2}. That was more than the value determined by the US Department of Energy, (1 μAcm^{-2}). The results of the Interfacial contact resistance and corrosion resistance of uncoated samples showed that Inconel 625 alloy for using in fuel cell bipolar plates the coating is necessary. As shown in the graph, the Andy category for the uncovered sample consists of two regions are taken in the active regions. Then, in the passive area an oxide layer is formed on the surface that protect against the corrosive environment.

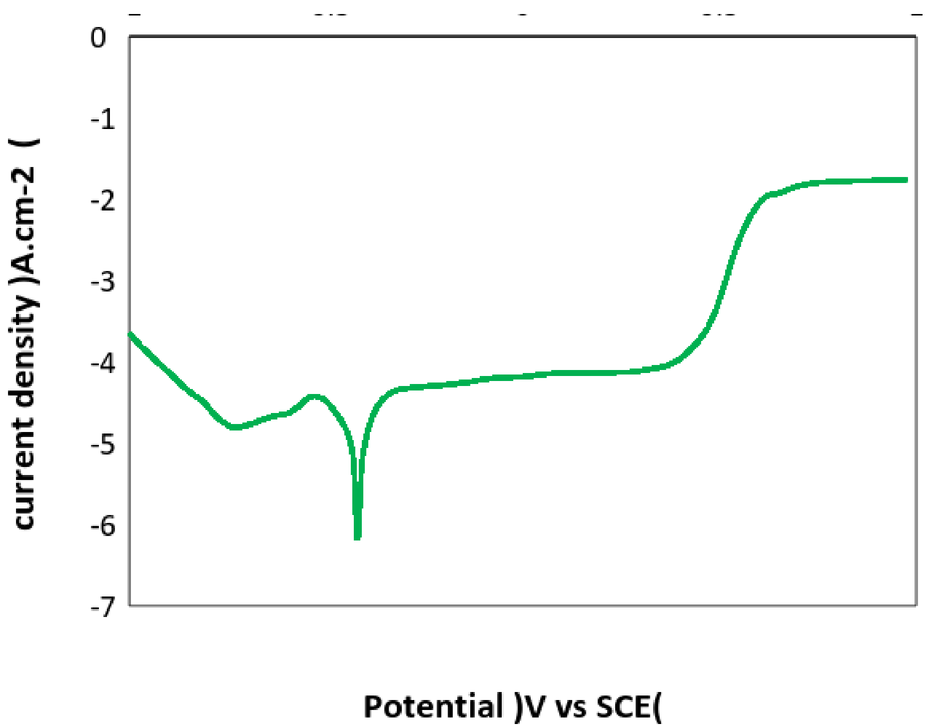


7. Check corrosion uncoated samples.

3.3.2. Corrosion behavior of TiN using Cathodic arc evaporation method

The results of corrosion behavior of TiN showed that corrosion potential and corrosion current became better that before. The samples covered with branches cathode became wider (Figure 8). In other words, resistance to corrosion is increased. The created area are as follows:

Figure 8.



Corrosion samples coated with TiN.

Area No. 1: the region is active. In this region with increasing potential, corrosion current increases sharply. And the pin holes in the coverage of TiN created.

Area No. 2: the region is passive. In this part by increasing the potential the current is stable. In other words, the non-conductive layer on the surface of the TiN coating, prevent further corrosion of coating which this layer may create because of the breakdown in TiN coating. According to the research of others [11], Ti(OH) or Ti(OH) will be made when placed on the surface prevent corrosion of the substrate.

Area No. 3: The active area is part of the graph that with increasing potential, corrosion current highly increased. In this area probably caused cavities on the surface of passive layers.

Area No. 4: In this area the passive oxide layer on the surface occur again. Considering that the protective oxide layer and covered, so Inconel substrate exposed to corrosive environments then the corrosive solution goes and makes cavities. But in this case, the oxide layer of Cr_2O_3 protects the substrate of Inconel.

The results of the coating cathodic arc evaporation method showed that all samples have improved in terms of corrosion and Interfacial contact resistance and the values were lower than those set by the US Department of Energy for the bipolar plates.

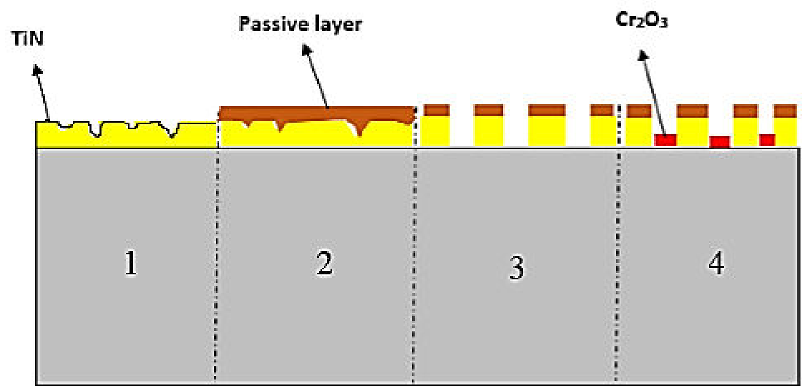


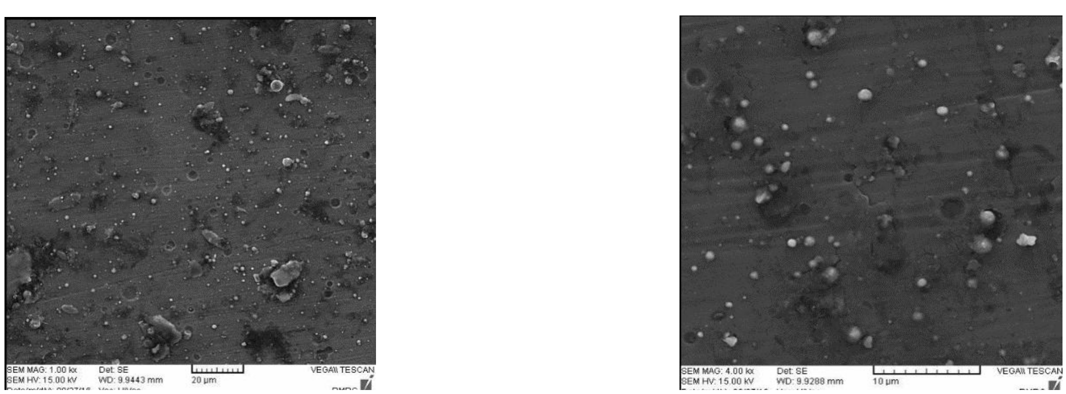
Figure 9, Schematic corrosion coating of TiN.

3.4. Scanning electron microscopy analysis

3.4.1. Surface morphology

As seen in the figures 10, there are holes in the surface of the sample, which can be caused by the coating method. Because the physical vapor deposition method causes the drilling of the surface.

Figure 10. The morphology of TiN coating prior to corrosion testing.



Base on the picture it is seen that cavities on the surface is coated with corrosion. Because the cavities are susceptible to corrosion and when exposed to corrosive environments are electrolytes in them will penetrate and cause corrosion hole.

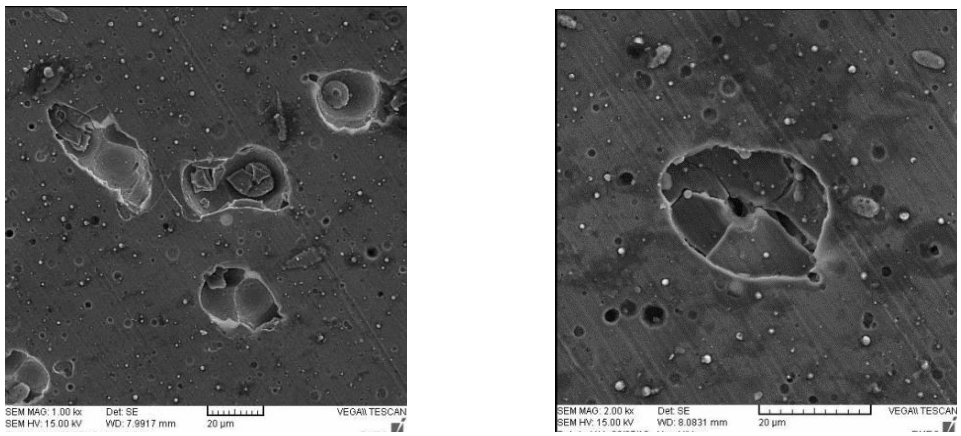


Figure 11. TiN coating surface morphology after corrosion test polarization.

3.4.2. The effect of coating thickness

Picture 12 are related to the cross-section before the corrosion test is carried out. In these images the cover and substrate are well defined.

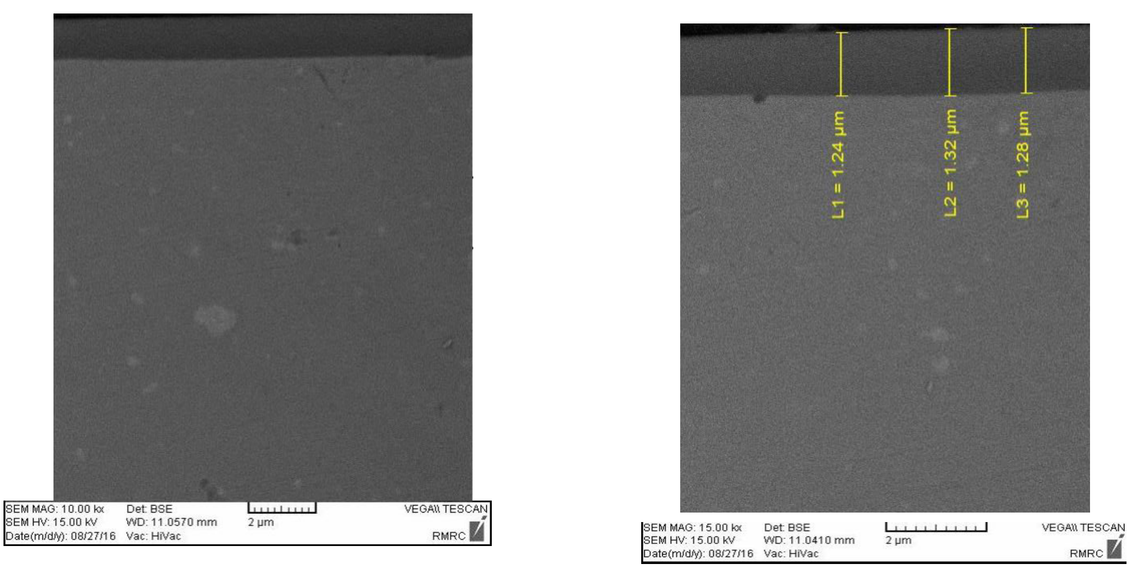


Figure 12. Typical cross-section with a thickness of 1200 nm TiN coating prior to corrosion testing.

Picture 13 shows the corrosion very well after the test. In Figure 1 it is seen that after testing, corrosion in some parts is corroded and disappeared. Figure 2 also shows is that the coating thickness is reduced.

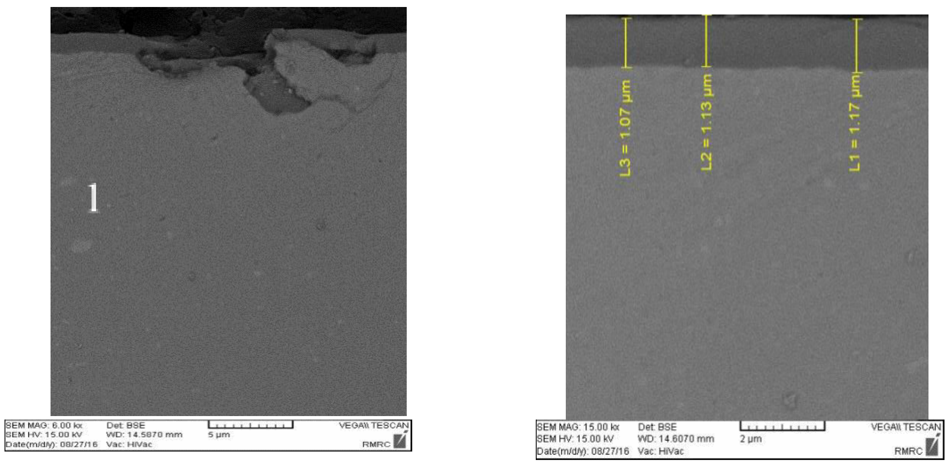


Figure 13. Typical cross-section with a thickness of 1200 nm TiN coating after the corrosion test.

4. Conclusion

1. The results showed that corrosion and Interfacial contact resistance of the Inconel alloy as the raw samples without coating is not suitable for using as bipolar plates. The results show that the oxidation furnace will increase the Interfacial contact resistance for this plate.

2. The results suggest that the more the contact resistance of the substrate surface is rougher, the less the Interfacial contact resistance appears. By increasing pressure the Interfacial contact resistance decreased.

3. By increasing the thickness of the coating TiN, Interfacial contact resistance first increased and then decreased. By increasing the coating thickness of TiN, corrosion current decreases.

4. Corrosion Testing and Interfacial contact resistance values obtained for all samples on the Coating, lower than those set by the US Department of Energy and for use in PEM fuel cell bipolar plates are appropriate.

5. The best sample in terms of corrosion and Interfacial contact resistance with the thickness of the coating sample 1200 nm, and roughness of the substrate 400 the Interfacial contact resistance

of the sample at about 8.14 mΩ.cm^2 and corrosion resistance for that is 0.1843 mΩ.cm^2. In this study, the sample as a good candidate for use in fuel cell bipolar plates is presented.

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