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Short communication

Testing and evaluation of aluminum coated bipolar plates of pem fuel cells operating at 70 °C

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Abstract

Corrosion resistant metal treated bipolar plates with higher rigidity and electrical conductivity than graphite were developed and tested for polymer electrolyte membrane (PEM) fuel cell applications. Four replicas of single cells were fabricated; two of graphite composites bipolar plates and the other two were coated aluminum. An E-TEK Series 14-W MEA with carbon cloth GDL, Nafion 112, 1.0 mg cm^{-2} platinum loading (anode and cathode) and 6.45 cm^2 active electrode areas, was fitted to each cell and operated under identical conditions. The obtained data from the two graphite cells were averaged and plotted and the other two aluminum cells' data were similarly treated and plotted on the same graph for comparison. Generally, the treated metallic bipolar plate provided at least a 22% savings in hydrogen consumption in comparison to graphite. This is attributed to the lower bulk and surface contact resistance of the coated aluminum plates used in this study in relation to graphite. The results of the lifetime testing conducted at 70 °C cell temperature under cyclic loading condition showed no indication of power degradation due to metal corrosion for at least 1000 h.

Keywords: Bipolar plate; PEM fuel cell

1. Introduction

As the oil and natural gas prices continue to increase to a level that threatens our national security the hydrogen economy seems to be the only alternative solution to this national problematic energy situation. Fuel cells are the main pillar of the hydrogen economy as they provide clean and reliable energy source and maintain healthy environment. Polymer electrolyte membrane (PEM) offers relatively high efficiency and power density as the main candidate to replace the internal combustion engine in the transportation applications. PEM stands among other types of fuel cells as a suitable source of power for combined automotive/stationary applications because of its relatively quick startup and shutdown operation as well as high efficiency and power density. Despite the fact that PEM fuel cells have an excellent potential to meet the economical and technical targets, considerable challenges are still facing the PEM industry as summarized in the following:

(1) Relatively expensive fabrication and material costs of the two main highly repeated components in the PEM fuel cell technology namely, bipolar plates and membrane electrode assembly (MEA).
(2) Durability of bipolar plates and MEA under the harsh corrosive environment and the electrochemical reaction inside the fuel cell.
(3) Startup, operation and shutdown processes in subfreezing condition.

The improvement of the cost effectiveness, durability and performance of these components will allow PEM fuel cells to penetrate the commercial market and compete with other energy sources. Accordingly, for the last two decades both industry and academia have been attempting to develop durable, efficient, cost effective MEA(s) and high corrosion resistance bipolar plates. The bipolar plates perform as the current conductors between

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cells, provide conduits for reactant gases flow, facilitate water and thermal management through the cell and constitute the backbone of the power stack. Gold coated titanium and niobium were the materials used by General Electric in the 1960s [1], however, they were replaced by graphite composites to reduce cost and weight. Nobel metals such as gold and platinum performed very similar to poco graphite bipolar plates [2] and in some cases showed more superior performance, however, the high cost of these metals has prohibited their utilization on the commercial scale.

Currently, graphite bipolar plates are considered the industry standard because of their high corrosion resistance and relatively low surface contact resistance. However, due to the graphite's brittleness, high permeability to gases and relatively expensive high volume manufacturing cost, alternative non-precious metals were tested by various researchers working in the field. Hermann et al. [3] reported that aluminum, stainless steel, titanium and nickel bipolar plates exposed to an operating environment similar to that of a fuel cell with a pH of 2–3 at temperatures of around 80 °C are prone to corrosion and dissolution. Moreover, a corrosion layer on the surface of a bipolar plate increases the electrical resistance and decreases the output of the cell. While this surface oxide layer protect the metal and stops the corrosion from progressing further through the lower layers beneath the surface, it forms, however, an electrically insulating interfacial layer. As the thickness of the oxide layer increases the electrical surface contact resistance also increases that accordingly cause a decrease in the electric power. Wang and Turner [4] studied various stainless steel samples, and noted the formation of passive films on AISI446 are mainly chromium oxide (Cr_2O_3), and the iron oxides play only a minor role. The passive film formed in the simulated PEMFC cathode environment is thicker than that formed in the simulated PEMFC anode environment, and the former has resulted in a higher interfacial contact resistance than the latter.

Therefore, these metals with their current situation were found unsuitable for PEM application due to their surface oxide or passive layer's electrical insulation property that does not meet the DOE target. Moreover, uncoated metal ions and oxides could directly foul the electrolyte and tarnish the catalyst in the membrane electrode assembly of the fuel cell that causes considerable adverse effects on the cell performance. Specifically, as non-protected metal bipolar plates are exposed to the harsh operating environment inside the fuel-cell which is very conducive to corrosion with relative humidity of more than 90%, acidity of 2–3 pH and temperature range of 60–80 °C, metal dissolution will occur. The dissolved metal ions diffuse into the membrane and then get trapped in the ion exchange cites inside the ionmer, resulting in lowering its ionic conductivity and causing considerable power degradation as described by Mehta and Cooper [5]. Accordingly, highly conductive corrosion resistance coatings with high bonding strength at the interfacial layer between the base metal substrate and the coating layer is required.

However, materials such as gold and phosphorous nickel show very high resistance to electrochemical corrosion, comparable to graphite, the traditional bipolar plate material. Gold plated stainless steel, on the other hand, showed great promise but lacked economic viability and exhibited poor bonding strength with the substrate. Major concerns have been the extent of corrosion (and its products) and the contact resistance of the surface passivation film. Candidate SSs have been tested and used as bipolar plates by different researchers. To avoid corrosion, metallic bipolar plates are coated with protective coating layers. Coatings should be conductive and adhere to the base metal without exposing the substrate to the corrosive media. Two types of coatings were reported [5–7] and investigated namely, carbon-based and metal-based coating materials for PEM fuel cell bipolar plates. Carbon-based coatings include: (1) graphite, (2) conductive polymer, (3) diamond like carbon and (4) organic self-assembled monopolymers. Metal-based coatings include: (5) noble metals, (6) metal nitrides and (7) metal carbides.

Woodman et al. [7] concluded that the coefficient of thermal expansion (CTE), corrosion resistance of coating and micropores and micro-cracks play a vital role in protecting the bipolar plate from the hostile PEM fuel cell environment. They also stated that even though PEM fuel cells typically operate at temperatures less than 100 °C, vehicle service would impose frequent startup and shut down conditions, and temperature differentials of 75–125 °C would be expected. A large difference in the CTE of the substrate and coating materials may lead to coating layer failure. One technique to minimize the CTE differential is to add intermediate coating layers with CTEs between that of adjacent layers. Also they added that materials such as aluminum, copper, nickel and tin are very susceptible to electrochemical corrosion in acidic solutions, typical of PEMFC operating conditions. However, materials such as gold and phosphorous nickel show very high resistance to electrochemical corrosion, comparable to graphite, the traditional bipolar plate material.

Li et al. [8] investigated the corrosion behavior of TiN coated type 316 stainless steel (SS). Their results revealed that TiN coating improved the corrosion resistance and electric conductivity of the SS 316. They also indicated that further efforts to improve the coating quality and to evaluate the long-term stability of 316SS/TiN coating systems under simulated conditions are still required.

Similar work was conducted by Cho et al. [9], they observed significant improvement in the lifetime of AISI 316 stainless steel bipolar plates as a result of coating them with corrosion-protective TiN layer. The authors have also mentioned that bipolar plates with low surface energy, low water contact angle >90° and high surface wetability could directly contribute to the flooding of the cathode side of the fuel cell. To evaluate surface energy of different materials, the water contact angle could be measured and evaluated. High contact angle is an indication of a high surface energy or surface tension of a material and low surface wetability. For instance water contact angle of TiN-coated 316 was almost the same as graphite and equal to 90° this represent an improvement of 316 stainless steel's contact angle that was 60°. Taniguchi and Yasuda [10] reported that very low water wetability of gas flow channels particularly where the condensed water inside the fuel cell tends to accumulate, caused noticeable increase in peak power of the fuel cell. Water accumulation or flooding of the flow conduits and gas diffusion layer

(GDL) will hinder the flow of the reactant gases and isolate the catalyst due to submersion resulting in considerable power degradation.

Lee et al. [11] applied physical vapor deposition (PVD) coating of YZU001 like diamond film on the 5052 aluminum alloy and SS 316L stainless steel and compared their performance with that of the graphite. The corrosion rates were determined by Tafel-extrapolation method from the polarization curves. The coated aluminum, 316L stainless steel and graphite were fabricated into single cell to measure contact resistance and to test cell performance. The results indicated that metallic bipolar plates, PVD coated 5052 aluminum and SS 316L, performed better than the graphite material at the low voltage but experienced shorter cell life. They also reported that the SS 316L plate with its naturally formed passive film had better corrosion rate than the YZU001 coated aluminum plate. However, the SS contact resistance was higher thus reducing its single cell performance. The Al-coated plates had better contact resistance and single cell performance, however, the cell life was shorter.

Brady et al. [12] have recently developed a preferential thermal nitridation process to form defect-free coatings or a pinhole free CrN/Cr_2N coating on a Ni–Cr alloy base plate, which show promise with excellent corrosion resistance and negligible contact resistance. They pointed out that dense, electrically conductive, corrosion-resistant Cr-nitride surfaces can be formed on Ni–Cr and Ni(Fe)–Cr base alloys at Cr levels <35 wt.% by thermal nitridation. They added that nitridation of 446 stainless steel (and likely other Fe–Cr base alloys), under certain conditions that modify the native passive oxide layer but do not form a dense Cr-nitride surface, can lower ICR by over an order of magnitude without compromising corrosion resistance. Both of these surface modifications show promise for protecting metallic bipolar plates in PEMFC environments, but this process cost poses a reason for concern.

Joseph et al. [13] electrochemically coated 304 stainless steel with conducting polymers polyaniline (PANI) and polypyrrole (PPY), showed improved corrosion resistance with acceptable contact resistance. Cost, durability and volume production were not mentioned in this study.

Therefore, the current study focused on a comparison between the effects of coated aluminum and graphite bipolar plates on hydrogen consumption, fuel cell efficiency and durability over wide range of power density output.

2. Experimental work

The experimental set-up consisted of four fuel cells encompassed in a hydrogen safety enclosure with a negative pressure test station connected to data acquisition system (DASYLab 5.6 software). All fuel cells output and operating parameters like current, voltage and power as well as temperature and reactant gases volume flow rate were recorded by the data acquisition system. The test station provided the reactants (hydrogen and air) and controlled the electric load while the data acquisition system measured and recorded the information. Both air and hydrogen are regulated by volume flow meters (Type FMA-A2300, Omega). The fuel cells are connected to a programmable electronic load (MCL488 DYNALoad) that was used in increments of constant current mode.

Electrode membrane assemblies, with 6.45 cm^2 active electrode area were loaded into four replicas of single fuel cells. Two of which were fabricated of graphite composites bipolar plates, and the other two cells were made of aluminum plates coated with high corrosion resistance, low interface and bulk contact resistance carbide-based patent pending alloy.

All cells were operated under identical conditions of controlled temperature at 70 °C, relative humidity at 95%, air flow rate of 470 $cm^3\ min^{-1}$ with back pressure of 7.5 psig and hydrogen pressure of 10 psig. The hydrogen was dead-ended at the exhaust manifold for all cells. The obtained data from the two graphite cells were averaged and plotted and other two metal cells data were similarly treated and plotted on the same graph for comparison. Each single cell text fixture consisted of two bipolar plates that contained a serpentine of rib channel patterns to allow the passage of hydrogen and air to the anode and cathode, respectively. The electrode membrane assemblies were acquired from (E-TEK, DeNora.), with double-sided Electrodes, Series 14-W MEA with carbon cloth GDL, Nafion 112, 1.0 mg cm^{-2} platinum loading (anode and cathode). The fuel cell operated with ambient air obtained from an industrial compressor and dry industrial grade hydrogen supplied by a metal hydride storage tank.

3. Results and discussion

The experimental results obtained from the two graphite plates were averaged and plotted in one single plot and labeled "graphite" as shown in the following figures. Also, the two coated aluminum bipolar plates were averaged, plotted and labeled as treated metal.

Fig. 1 shows the average polarization and power density curves for the first pair of single aluminum coated fuel cells. Also, the results obtained from the second pair of single fuel cells made of composite graphite were averaged and plotted. The results exhibited better performance of the aluminum coated bipolar plate in comparison with the graphite. For example, at a 200 mA cm^{-2} current density the cell voltage outputs were 0.70 and 0.55 V for treated metal and composite graphite bipolar plate, respectively. Also, Fig. 1 depicts that the maximum average output power density was 0.32 and 0.14 W cm^{-2} for treated

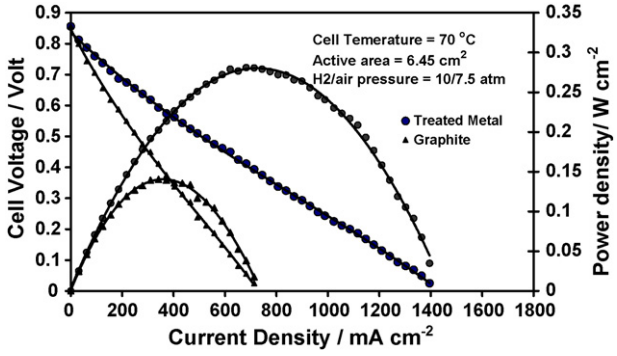

Fig. 1. Polarization curve and power density.

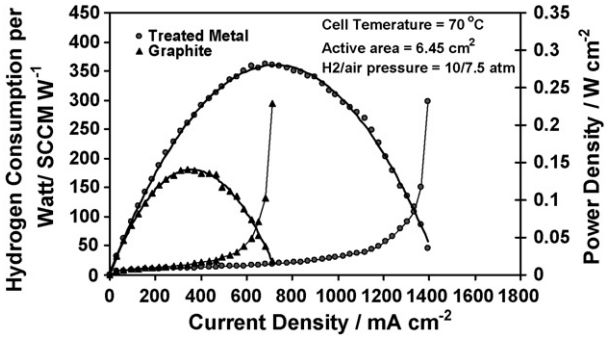

Fig. 2. Hydrogen consumption and power density.

metal and composite graphite, respectively. This is attributed to the lower bulk and contact resistances of metal bipolar plate compared to graphite. Moreover, it was found that the graphite cell resistance is approximately 2.5 times higher than the metallic cell under the same operating conditions.

Fig. 2 exhibits the distribution of power density and hydrogen consumption per watt versus current density for both coated metal and composite graphite bipolar plates. The results showed that the hydrogen consumption per watt using metal bipolar plate is lower than graphite. For example, the hydrogen consumption per watt at a current density of 200 mA cm^{-2}; was 10.4 and 13.1 cm^3 min^{-1} W^{-1} when coated metal and composite graphite were used as bipolar plates, respectively.

Preliminary experimental results measured showed at least 22% savings in hydrogen consumption because of the lower bulk and contact resistance of metal than graphite. A simple cost analysis of electric energy losses as heat due to the bulk resistance of aluminum and graphite showed that aluminum bipolar plates save electric energy from converting to heat in the amount of US$ 1060 per year for a 500 kW unit.

The power density and fuel cell average efficiency distributions for comparing metal and graphite bipolar plate performances at different levels of current density are depicted in Fig. 3. The figure shows that the efficiency of the fuel cell using treated aluminum is higher than graphite. For example, the efficiency of fuel cell at 200 mA cm^{-2} was 58% and 45% when using metal and composite graphite as bipolar plate, respectively.

Fig. 4 shows the polarization curves and power density measured at different time intervals namely, 30 and 900 working

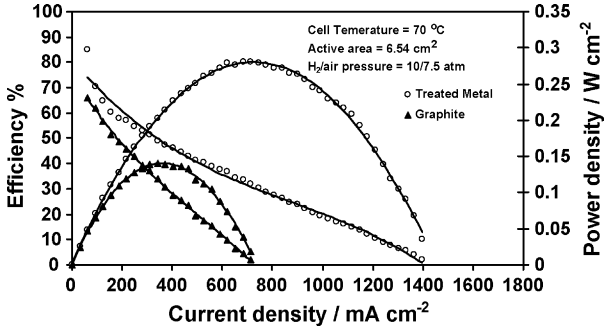

Fig. 3. Efficiency and power density.

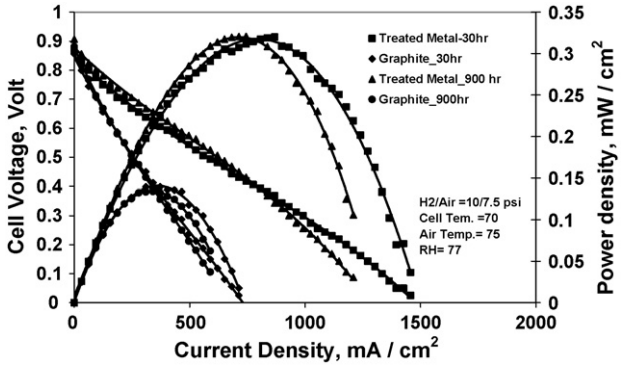

Fig. 4. Polarization and power curves through 1000 h of operation for treated metal and graphite after 30 and 900 h.

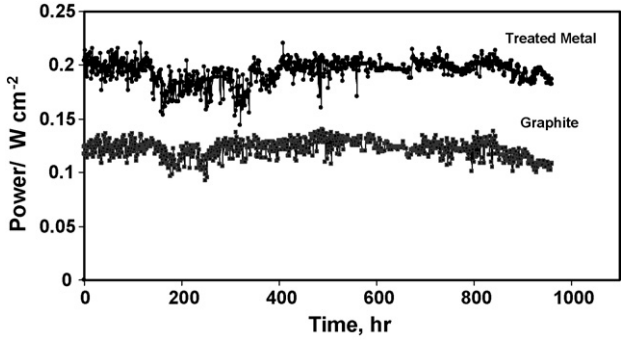

Fig. 5. Life time test of treated metal bipolar plates and graphite bipolar plate operated under cyclic loading at 70 °C.

hours for treated metal and graphite. No noticeable deviation was observed and the maximum percentage difference was 0.33%.

The four fuel cells were tested under the same operating conditions and each cell powered the exact variable loading. The metallic bipolar plates performed at 70 °C for approximately 1000 h without a sign of power degradation due to corrosion. The average output power of the pair of coated aluminum fuel cells were averaged and plotted in Fig. 5. Similarly, the other two graphite fuel cells were average and statistically treated and plotted on the same graph for comparison.

The parallel and very similar performance trends of graphite and coated aluminum provide a clear proof that no power degradation was caused by metal corrosion. Graphite is known to be non-corrosive and therefore it can be used as a reference of comparison.

In this study, industrial hydrogen and oxidant air were used. Air was provided by industrial compressors. Oil, particles and impurities were not effectively filtered from the industrial compressors and hydrogen tanks before feeding the fuel cells. This did not cause any fouling of the MEA or any power degradation as could be observed in Fig. 3. Also there is no sign of corrosion on the metallic bipolar plate after 1000 h operation as depicted in Fig. 5.

4. Conclusions

The results shown above indicate that treated metal bipolar plates can be used in PEM fuel cells because they have

a higher performance than graphite. Aluminum coated bipolar plates showed a 22% saving in hydrogen consumption, higher efficiency and durability in relation to graphite. The coated aluminum bipolar plates performed for 1000 h at 70 °C without any sign of power degradation due to corrosion. However, accelerated corrosion testing needs to be conducted and longer duration for the lifetime testing is required to confirm the durability and efficiency of both MEA and bipolar plates.

In addition, metallic bipolar plates are noted for their ductility and lack of brittleness that plagues the graphite plates and causes cracking and mechanical failure under the stack tightening force. Accordingly, metallic plates are deemed safer, more robust and more reliable than the graphite plates.

References

[1] P. Costamagna, S. Srinivasan, J. Power Sources 102 (2001) 253–269.
[2] J. Wind, R. Spah, W. Kaiser, G. Bohm, J. Power Sources 105 (2002) 256–260.
[3] A. Hermann, T. Chaudhuri, P. Spagnol, Int. J. Hydrogen Energy 30 (2005) 1297–1302.
[4] H. Wang, J.A. Turner, J. Power Sources 128 (2004) 193–200.
[5] V. Mehta, J.S. Cooper, J. Power Sources 144 (2003) 32–53.
[6] R.L. Borup, N.E. Vanderborgh, Mater. Res. Soc. Symp. Proc. 393 (1995) 151–155.
[7] A.S. Woodman, E.B. Anderson, K.D. Jayne, M.C. Kimble, Development of Corrosion-Resistant Coatings for Fuel Cell Bipolar Plates, American Electroplaters and Surface Finishers Society 1999, AESF SUR/FIN'99 Proceedings, vol. 6, 1999, pp. 21–24.
[8] M. Li, S. Luo, C. Zeng, J. Shen, H. Lin, C. Cao, Corros. Sci. 46 (2004) 1369–1380.
[9] E.A. Cho, U.-S. Jeon, S.-A. Hong, I.-H. Oh, S.-G. Kang, J. Power Sources 142 (2005) 177–183.
[10] A. Taniguchi, K. Yasuda, J. Power Sources 141 (1) (2005) 8–12.
[11] S.-J. Lee, C.-H. Huang, J.-J. Lai, Y.-P. Chen, J. Power Sources 131 (2004) 162–168.
[12] M.P. Brady, P.F. Tortorelli, K.L. More, H.M. Meyer, III, L.R. Walker, H. Wang, J.A. Turner, B. Yang, R.A. Buchanan, Cost-Effective Surface Modification for Metallic Bipolar Plates, DOE FY 2004 Progress Report.
[13] S. Joseph, J.C. McClure, R. Chianelli, P. Pich, P.J. Sebastian, Int. J. Hydrogen Energy 30 (2005) 1339–1344.