# Proton Exchange Membrane Fuel Cell Using Membrane Electrode Assembly Based on Platinum-Carbon Electrocatalyst with Activated Carbon-Chitosan-Nickel

Made Sucipta\*<sup>‡</sup>, James Sibarani<sup>\*\*</sup>, I Gede Nengah Wika Gunawan<sup>\*</sup>, Satriyo Galang Bhuana Putra<sup>\*</sup>, Rivaldo Anderson Robert<sup>\*</sup>, Shaquille Kandou<sup>\*</sup>, Putu Brahmanda Sudarsana<sup>\*</sup>

\*Department of Mechanical Engineering, Faculty of Engineering, Universitas Udayana, 80361 Badung, Bali \*\*Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Udayana, 80361 Badung, Bali

> Corresponding Author: Made Sucipta, 80361, m.sucipta@unud.ac.id Received: 30.09.2022 Accepted: 18.11.2022

Abstract- The development of proton exchange membrane fuel cells (PEMFC) has been focused on membrane electrode assembly (MEA). In this research, MEA based on Platinum-Carbon electrocatalyst (Pt/C) mixed with activated carbon-chitosannickel, has been fabricated and tested on a PEMFC prototype. As a comparison, two commercial MEAs based on the Nafion membrane were also tested with Pt/C. It is found that the highest voltage achieved from the developed MEA without loading is 0.43 V, lower than the voltage achieved by the commercial MEA. When an electrical resistance load was given, a decrease in the electrical voltage was observed. However, the developed MEA is able to produce the largest power density value at an electric current density greater than 2 mA/cm<sup>2</sup>. The developed MEA has shown sufficient performance when compared to commercial MEA on the market. This preliminary research opens the potential for MEA development using more abundant and lower-cost materials, such as the activated carbon-chitosan-nickel.

Keywords Membrane Electrode Assembly; PEM Fuel Cell; Chitosan; Nickel; Electrocatalyst.

#### 1. Introduction

Fuel cells are identified as promising electrical energy generators that can provide clean and efficient energy for stationary, transportation, and portable power applications in the 21st century [1], [2], [3]. The electrical energy generated from these fuel cells is obtained through a continuous electrochemical process in converting chemical energy fuels (such as hydrogen, natural gas, methanol, ethanol, and other gases) into electrical energy (and some heat and water vapor) during the fuel cell cycle when oxidant is supplied [4]. The type of fuel cell that can be used in mobile/portable applications is the Proton Exchange Membrane Fuel cell (PEMFC). The hallmark of PEMFC compared to other types of fuel cells is the presence of a Membrane Electrode Assembly (MEA) section. MEA serves as a separator between the anode and cathode, each of which is a place for supplying hydrogen and oxygen flows. Another function is as a distributor of protons or positive charges from hydrogen to the cathode until it finally merges with a negative charge and oxygen which leads to water products. Recently, the development of PEMFC technology has focused more on the synthesis of membranes into MEA [5].

In addition to the use of nafion which has been widely used as a membrane-making material, several studies have begun to examine the manufacture of membranes using organic materials, one of which is by using chitosan [6]. However, the use of chitosan has not been widespread, this can be due to its low thermal conductivity [5]. Very low thermal conductivity can inhibit H+ ions to react with oxygen, so to increase the thermal conductivity, chitosan can be mixed with metal or organic materials, one of which is using activated carbon [7], [8].

The catalyst materials that have been developed so far are based on carbon and metals such as platinum (Pt) [9]. Seeing the advantages of activated carbon compared to ordinary carbon, especially in terms of a wider surface area and high porosity, which will allow for better hydrogen ion movement, research on the use of activated carbon as an ingredient in the

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manufacture of catalysts has begun to be considered. The use of Platinum, which is a fairly expensive material, greatly affects the marketing of PEMFC. In fact, on this earth there are light metal materials with almost the same character as platinum so that they can be used as a substitute for platinum, such as nickel for example. Nickel has similar characteristic which is their ability to bind oxygen although too strong [10]. Other than that, Nickel has good corrosion resistance which is fit as electrocatalyst on the cathode where the oxygen reduction reaction [11].

Evaluating from the maturity of the technology and materials used as the base material for the membrane, there are already several MEAs that have been produced, either in the demo stage of use or commercialization. The commercialized MEA base material mostly uses a mixture of platinum and carbon (Pt/C) which is supported by several other materials as complementary components. However, long term use of MEA will reduce its performance so that it will reduce the output power when applied to PEMFC. Based on the ongoing development of research on MEA and the commercialization of MEA which in its application will experience performance degradation, it is necessary to try to gradually substitute the use of Pt/C by using other materials as substitutes such as nickel. To determine the performance of the developed MEA, an MEA testing tool has been designed which is applied to PEMFC. As a comparison, several existing commercial MEAs will be tested.

# 2. Materials and Methods

The design of the MEA testing apparatus on the PEMFC consists of a DC power supply, an electrolyzer unit, and a PEMFC unit which can be disassembled to place the MEA as the subject of tests. The working principle of this test system is to adjust the DC power supply to produce an output voltage in the range of 0-4 Volts. The electrolyzer was given distilled water. From this electrolyzer there will be a separation reaction of water (H<sub>2</sub>O) into hydrogen (H<sub>2</sub>) and oxygen (O<sub>2</sub>). Hydrogen produced from this electrolyzer is flowed on the anode side of the PEMFC and on the cathode side of the oxygen supply using the availability of atmospheric air in the test environment. The schematic of the MEA test equipment design on PEMFC can be seen in Figure 1. Figure 2 shows the results of the MEA test equipment design on PEMFC.



Fig. 1. Visualization of the MEA testing tool design on PEMFC

The PEMFC section has been installed with MEA which has sides as anode and cathode. The structure of PEMFC is shown in Figure 3, which consists of the cathode and anode end plate, current collector, graphite, gasket layer, gas diffusion later, and the MEA. Hydrogen passing through the anode meets MEA will turn into a proton ( $H^+$ ). The ability of

proton permeability will meet with oxygen on the cathode side which will produce electrons that will generate electricity, in addition to  $H_2O$  as a result of chemical reactions. The electron collector in the PEMFC will be connected to the resulting electric voltage meter. By giving a certain load, the electric current generated will also be obtained.



Fig. 2. MEA testing equipment on PEMFC



Fig. 3. PEMFC Structure

The process of making a membrane electrode assembly (MEA) based on activated carbon-chitosan-light metal is carried out with several preparations such as procurement of chitosan, carbonized bamboo and physically activated. In addition, the manufacture of catalysts using Pt/C catalysts mixed with light metal materials, such as the use of nickel. several supporting materials such as acetic acid, isoproponal, adhesives, carbon paper and other supporting tools for the manufacturing process.

The manufacture of activated carbon is carried out by the carbonization process at a temperature of 650°C with a holding time of 2 hours at that temperature. Furthermore, after filtering at a certain mesh size, activation is carried out at 625°C, each with the same holding time as in the carbonization process, which is for 2 hours.

Activated carbon that is ready to use is mixed with chitosan with a certain mass, then given acetic acid solvent, stirred using a stirrer for 2 hours. Then it is poured into the prepared glass mold. The natural drying process is carried out by waiting for about 3 days at atmospheric temperature. Furthermore, the membrane is cut to size according to the MEA size that has been determined on the PEMFC prototype. The catalyst was prepared by mixing 40% Pt/C with a light metal material, namely nickel. Furthermore, with the painting

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method, the catalyst is printed on both sides of the membrane. The outermost layer uses carbon paper to function as a gas diffusion layer (GDL). The hot press method was applied to MEA with a temperature of around  $200^{\circ}$ C and a mass loading of 2 kg.

The prepared MEA based on activated carbon-chitosanlight metal (nickel) (MEA-L) was then tested on a PEMFC prototype. As a comparison, two commercial MEA (MEA-K and MEA-U) based on Naphion membrane were also tested, with platinum-carbon catalyst. The flow of fuel using hydrogen gas is regulated with a certain mass flow rate which is assumed to be stable as a result of electrolysis of distilled water, while the need for oxygen is by using atmospheric air flow. The condition of the test room is assumed to be in standard conditions (STP). The load on the generated electricity ranges from 0 -300 ohms. The recording of voltage and electric current is done after the measuring device (AVOmeter) shows a stable number.

#### 3. Result and Discussion

The results of the electrical voltage test produced in this study reached the highest value of 0.81 V at no load for MEA-U. The load is given starting from a small load starting from 3.4 ohms to the highest being given a resistance of 300 ohms to obtain the electric current generated. The voltage – current diagram of the MEA's performance can be shown in Figure 4. The greater the electric current, the lower the voltage obtained. As shown in Figure 4, for MEA-U at an electric current of 54.2 mA, an electrical voltage of 0.18 V is obtained. This is due to some total losses experienced from losses in activation voltage, ohmic voltage, and load transportation [12], [13].

The resulting current density and power density can be described as shown in Figure 5. As shown in Figure 5, that the maximum power density in the MEA-U test produces 0.86 mW/cm2 at an electric current density of 2.33 mA/cm2. This parameter indicates the size of the MEA required for the PEMFC to produce a certain amount of electrical power. From Figure 4 it can also be seen that there is a maximum power that can be achieved by an MEA in this PEMFC application.

Interestingly, all the test results obtained showed a good trend of conformity with the results of studies from other researchers that have been published [12]-[18]. The factors that cause a slight difference in the acquisition of power density and electric current density are the magnitude of the contact pressure distribution on the surface of the PEMFC, the reactant flow rate to the PEMFC inlet channel, the composition of the MEA, the magnitude of the catalyst loading, and the dimensions of the part of the PEMFC itself. The pressure distribution that occurs in the PEMFC depends on the number of bolt connections used by the PEMFC. The more bolt connections, of course, the increase in pressure distribution leads to a decrease in ohmic contact resistance, resulting in an increase in output power [17]. The flow rate of hydrogen gas also has an effect on electrochemical reactions that lead to the power density and the resulting electric current density [16].



Fig. 4. Comparison graph of voltage-to-current between MEA-L, MEA-K, and MEA-U



Fig. 5. Comparison graph of power density-to-current density of MEA-L, MEA-K, and MEA-U

Dimensions in the design of PEMFC parts such as the bipolar plate design in PEMFC also affect the power and electric current delivered. Research on the design of a bipolar plate with a flow pattern that has many curvilinear indentations compared to sharp indentations is able to provide higher performance [18]. This is caused by the lack of vortex flow in the plate flow so that the gas flow does not experience a decrease in the velocity and flow rate of the reactants [19]. The greater the value of catalyst loading with Pt/C material also affects the performance of PEMFC because it has an effect in accelerating the reaction of separating positive and negative charges from hydrogen gas as a reactant [20]. Broadly speaking, the results of this test can illustrate that the design of the MEA testing system on the PEMFC that has been made can be used to determine the characterization of the MEA being tested.

According to the result, it appears that the MEA based on activated carbon-chitosan-light metal (MEA-L) has the same tendency for the results to be obtained as the commercial MEA. This shows that the characteristics of the MEA produced have shown their function well, although the results

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are still not optimal when compared to MEA-U, but compared to the commercial MEA-K, it has shown the equality of the results obtained. We can see that the highest electrical voltage that can be achieved by MEA-L is 0.43 V, while MEA-K reaches 0.51 V, all under test conditions without electrical load. Furthermore, when given an electrical load, the electric voltage on MEA-K drops quite drastically so that the magnitude of the electrical voltage obtained is almost the same as the electric voltage achieved by MEA-L with the increasing electric current generated. This shows that the electrical power generated will be almost the same, as confirmed in Figure 5. If we look in more detail, then the electric power density achieved by MEA-L shows the largest value, which is 0.67 mW/cm2 at an electric current density above 2.8. mA/cm2. From these results, it shows that MEA-L, which was developed from activated carbon-chitosan-nickel combined on a Pt/C catalyst, can give better results.

The current research gives a significant finding on the potentials of lower cost MEA in the application of PEMFC. Several papers have demonstrated fuel cell's potential implementation in various grid models through simulation [21]–[23]. Zafar [23] proposed a control model of the PEMFC implementation integrated with solar photovoltaic (PV) for a smart home design where it is capable of controlling the hydrogen production in PEMFC as a way to store excess energy from solar PV. With a lower price PEMFC design, indeed this scheme can be more feasible and sustainable in the future.

# 4. Conclusion

The MEA test prototype design on PEMFC in the present study can be used with similar trend results to the results of studies that have been carried out by other researchers so that it is feasible to use the MEA testing developed at this time. The development of MEA based on activated carbon-chitosan-light metal, especially in the use of nickel catalyst, has also been successfully tested. The results showed that the MEA made had shown good performance. The highest power densities are achieved at higher current densities. This is evident from the comparison that has also been tested using two existing commercial MEAs. Although it has not shown optimal performance, in other respects it also shows performance that is equivalent to one of the MEA tested. But another advantage that can be obtained is that the use of light metals, such as nickel, can reduce production costs using Pt/C catalysts which are still expensive at this time.

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# References

 M. Wang, Y. Ding, J. Hu, L. Xu, and X. Yang, Numerical simulation of water and heat transport in the cathode channel of a PEM fuel cell, *International Journal of Hydrogen Energy*, Vol. 47, No. 20, 2022: 11007–11027. https://doi.org/10.1016/j.ijhydene.2022.01.143

[2] Y. Yang, H. Jia, Z. Liu, N. Bai, X. Zhang, T. Cao, J. Zhang, P. Zhao, and X. He, Overall and local effects of operating parameters on water management and performance of open-cathode PEM fuel cells, *Applied Energy*, Vol. 315, 2022: 118978

[3] Y. Wang, D. F. Ruiz Diaz, K. S. Chen, Z. Wang, and X. C. Adroher, Materials, technological status, and fundamentals of PEM fuel cells – A review, *Materials Today*, Vol. 32, 2020: 178–203. https://doi.org/10.1016/j.mattod.2019.06.005

https://doi.org/10.1016/j.apenergy.2022.118978

- [4] L. Zhang, S. R. Chae, Z. Hendren, J. S. Park, and M. R. Wiesner, Recent advances in proton exchange membranes for fuel cell applications, *Chemical Engineering Journal*, (204–205), 2012: 87–97. https://doi.org/10.1016/j.cej.2012.07.103
- Y. Wang, K. S. Chen, J. Mishler, S. C. Cho, and X. C. Adroher, A review of polymer electrolyte membrane fuel cells: Technology, applications, and needs on fundamental research, *Applied Energy*, Vol. 88, No. 4, 2011: 981–1007. https://doi.org/10.1016/j.apenergy.2010.09.030
- [6] A. C. Humelnicu, P. Samoila, M. Asandulesa, C. Cojocaru, A. Bele, A. T. Marinoiu, A. Sacca, and V. Harabagiu, Chitosan-sulfated titania composite membranes with potential applications in fuel cell: Influence of cross-linker nature, *Polymers*, Vol. 12, No. 5, 2020. https://doi.org/10.3390/POLYM12051125
- [7] D. Chinnadurai, P. Karuppiah, S. M. Chen, H. J. Kim, and K. Prabakar, Metal-free multiporous carbon for electrochemical energy storage and electrocatalysis applications, *New Journal of Chemistry*, Vol. 43, No. 29, 2019: 11653–11659. https://doi.org/10.1039/c9nj01875a
- [8] X. Jia, Q. Li, C. Ao, R. Hu, T. Xia, Z. Xue, Q. Wang, X. Deng, W. Zhang, and C. Lu, High thermal conductive shape-stabilized phase change materials of polyethylene glycol/boron nitride@chitosan composites for thermal energy storage, *Composites Part A: Applied Science and Manufacturing*, Vol. 129, 2020: 105710. https://doi.org/10.1016/j.compositesa.2019.105710
- [9] P. C. Okonkwo, O. O. Ige, E. M. Barhoumi, P. C. Uzoma, W. Emori, A. Benamor, and A. M. Abdullah, Platinum degradation mechanisms in proton exchange membrane fuel cell (PEMFC) system: A review, *International Journal of Hydrogen Energy*, Vol. 46, No. 29, 2021: 15850–15865.

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https://doi.org/10.1016/j.ijhydene.2021.02.078

- O. T. Holton and J. W. Stevenson, The Role of Platinum in Proton Exchange Membrane Fuel Cells Evaluation of platinum's unique properties for use in both the anode and cathode of a proton exchange membrane fuel cell, *Platinum Metals Rev*, Vol. 57, No. 4, 2013: 259–271. http://dx.doi.org/10.1595/147106713X671222
- [11] T. Jin, W. Zhang, N. Li, X. Liu, L. Han, and W. Dai, Surface characterization and corrosion behavior of 90/10 copper-nickel alloy in marine environment, *Materials*, Vol. 12, No. 11, 2019. https://doi.org/10.3390/ma12111869
- [12] K. Benmouiza and A. Cheknane, Analysis of proton exchange membrane fuel cells voltage drops for different operating parameters, *International Journal* of Hydrogen Energy, Vol. 43, No. 6, 2018: 3512– 3519. https://doi.org/10.1016/j.ijhydene.2017.06.082.
- U. Rost, C. Mutascu, J. Roth, C. Sagewka, and M. Brodmann, Proof of Concept of a Novel PEM Fuel Cell Stack Design with Hydraulic Compression, *Journal of Energy and Power Engineering*, Vol. 9, No. 9, 2015: 775–783. https://doi.org/10.17265/1934-8975/2015.09.003
- W. R. Chang, J. J. Hwang, F. B. Weng, and S. H. Chan, Effect of clamping pressure on the performance of a PEM fuel cell, *Journal of Power Sources*, Vol. 166, No. 1, 2007: 149–154. https://doi.org/10.1016/j.jpowsour.2007.01.015
- [15] P. Scott, R. K. Calay, and Y. K. Chen, Experimental evaluation into novel, low cost, modular PEMFC stack, *Energy Procedia*, Vol. 29, 2012: 567–575. https://doi.org/10.1016/j.egypro.2012.09.066
- [16] A. Syampurwadi, H. Onggo, Indriyati, and R. Yudianti, Performance of PEM fuel cells stack as affected by number of cell and gas flow-rate, *IOP Conference Series: Earth and Environmental Science*, 60, 2017: 012029. https://doi.org/10.1088/1742-6596/755/1/011001

- C. Y. Wen, Y. S. Lin, and C. H. Lu, Experimental study of clamping effects on the performances of a single proton exchange membrane fuel cell and a 10-cell stack, *Journal of Power Sources*, Vol. 192, No. 2, 2009: 475–485. https://doi.org/10.1016/j.jpowsour.2009.03.058
- [18] M. E. S. Youssef, R. S. Amin, and K. M. El-Khatib, Development and performance analysis of PEMFC stack based on bipolar plates fabricated employing different designs, *Arabian Journal of Chemistry*, Vol. 11, No. 5, 2018: 609–614. https://doi.org/10.1016/j.arabjc.2015.07.005
- [19] K. B. Hong, D. W. Kim, J. Kwark, J. S. Nam, and H. S. Ryou, Numerical study on the effect of the pipe groove height and pitch on the flow characteristics of corrugated pipe, *Energies*, Vol. 14, No. 9, 2021. https://doi.org/10.3390/en14092614
- [20] N. M. Zahari and A. A. Aziz, Effect of platinum catalyst loading on membrane electrode assembly (MEA) in proton exchange membrane fuel cell (PEMFC), 2012 10th IEEE International Conference on Semiconductor Electronics, ICSE 2012 -Proceedings, 2012: 669–673. https://doi.org/10.1109/SMElec.2012.6417232
- [21] G. Pourabedin and F. Ommi, Modeling and Performance Evaluation of Standalone Solid Oxide Fuel Cell for Aircraft APU-II: Dynamic Performance, *International Journal of Smart Grid*, Vol. 3, No. 1, 2019: 35–41. https://doi.org/10.20508/ijsmartgrid.v3i1.38.g44
- [22] S. Gherairi, Zero-Emission Hybrid Electric System: Estimated Speed to Priotize Energy Demand for Transport Applications, *International Journal of Smart Grid*, Vol. 3, No. 4, 2019: 180–187. https://doi.org/10.20508/ijsmartgrid.v3i4.76.g65
- B. Zafar, Design of a Renewable hybrid photovoltaic-Electrolyze-PEM/Fuel Cell System using Hydrogen Gas, *International Journal of Smart Grid*, Vol. 3, No. 4, 2019: 202–207. https://doi.org/10.20508/ijsmartgrid.v3i4.83.g68