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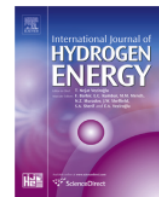
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ARTICLE INFO

ABSTRACT

The characterization of electrodes and performance tests for membrane electrode assemblies (MEAs) with varying Pt content have been carried out. The electrodes were made using a spraying method. The physical properties of the electrodes (i.e., surface structure, content and distribution of catalysts) were characterized using scanning electron microscope—energy-dispersive X-ray spectroscopy (SEM—EDX), and MEA performance tests were performed through a fuel cell test station with varying catalyst content, fuel cell temperature and with applied back pressure. The physical characterization of the electrode indicated that the Pt catalyst was uniformly dispersed on both the surface and cross section of the catalyst layer (CL), with a Pt loading between 7.5 and 21.5 wt%. The current—voltage performances generally increased with Pt content. Similarly, the use of back pressure and increasing the fuel cell operation temperature improved MEA performance.
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1. Introduction

Proton exchange membrane fuel cells (PEMFCs) are highly efficient fuel cells. PEMFCs produce electrical energy from an electrochemical reaction between hydrogen gas as the fuel and oxygen gas as the oxidant with a proton exchange membrane as the electrolyte and catalyst layers contained in both of the electrodes [1].

The most important component in the PEM fuel cell is the membrane electrode assembly (MEA). Therefore, many studies have been conducted on the manufacture of MEAs to develop an MEA with a high current density and long-term durability. MEA performance depends on many factors, including the type and thickness of the gas diffusion layer (GDL) [2], the electrode manufacturing process, the performance of CL and the MEA manufacturing method [3,4].

An MEA consists of two electrodes on either side of a polymeric electrolyte membrane. Three important processes occur in an MEA: proton transport from the membrane to the catalyst, electron transport from the current collector to the catalyst and vice versa and the transport of gas reactants and products to and from CL and gas channel [5,6].

The electrode structure consists of a gas diffusion media (GDM) or macroporous layer as backing layer, a microporous

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layer (MPL) and a CL. Combination of GDM or macroporous layer and MPL called GDL [7]. The electrodes must not only be sufficiently conductive but also hydrophobic enough. The function of hydrophobicity in electrode is to keep out the water and to enhance the water flow in order to avoid suspended water between the pores due to the adhesion force between water and carbon substrate.The importance of a hydrophobic layer able to increase the water drops removal tendency and prevent the risk of water flooding [8].

The electrode manufacturing processes have been studied through either the simulation or optimization of PEM fuel cell performance [9]. The studied manufacturing methods [10] include casting and brushing [9,11], electrospraying with a dual catalyst layer [12], thin layer [11], printing [13,14], decal [8], impregnation [15], spraying [2,12], CCM [4,16], and electrodeposition methods [17,18]. Su et al. [12] demonstrated the spraying method by spraying ink onto the electrolyte membrane while varying the catalyst content. Meanwhile, Su et al. [19] compared spraying methods with a double catalyst layer (DCL) with the conventional methods.

Among the above-mentioned electrode manufacturing processes, the method that involves spraying onto the GDL is simple and can be used to manufacture a high-performing electrode with a uniform distribution of the catalyst [20].

The common electrode surface structure was characterized using SEM–EDX [15]; the catalytic activity was observed using cyclic voltammetry and electrochemical impedance spectroscopy (EIS) [21,22].

The relationship between the catalyst content in carbon and the PEMFC performance has been widely studied [11,23]. The influence on the electrode performance of different platinum concentrations in the carbon (20, 40 and 60 wt%) in single cell stack was reported by Cho [23]. Cho found that increasing the concentration of Pt in the carbon does not directly correlate to improved performance. The performance at 60 wt% Pt decreased dramatically due to an increased mass transfer resistance and a decrease in the number of micropores that caused flooding.

In addition to the catalyst content, the thickness of the catalyst layer is also a subject of study. Utilizing a multi-catalyst layer (MCL) with different catalyst and Nafion loading in each layer is one strategy to improve catalyst efficiency, prevent flooding at the CL–GDL interface and increase mass transfer [19]. Srinivasarao [24] attempted to theoretically compare the performance of a single CL with a multi-layer CL at cathode by varying the ionomer content. Electrodes with a multi-layer CL and an ionomer content between 22 and 34 wt % exhibit good performance and better handling of water production. However, in contrast with Srinivasarao's results, the production of a multi-layer CL in this study is intended to reduce the interfacial resistance at the GDL on one side and the electrolyte membrane on the other side. Therefore, the CL is made up of three layers: the first layer is a mixture of Pt/C catalyst, PTFE and isopropanol as a solvent; the second layer is a mixture of Pt/C, Nafion solution and the solvent; and the third layer is a solution of Nafion as the primary constituent with isopropanol as a solvent.

Efforts have been made to improve the performance of the PEMFCs by optimizing the operating conditions. Santarelli [25] has attempted to improve the PEMFC performance through changes in the cell temperature and the pressure of the reactants. Optimization of the operating conditions is easier because changes can be made when the fuel cell running. The effects of channel depth, temperature and pressure have also been investigated [26], and the optimum performance is obtained at a depth of 1.5 mm for the anode and 1 mm for the cathode. In general, PEMFC performance increases with the increasing temperature and pressure caused the increasing kinetic energy of the reactants. However, obtaining consistent values for some kinetic parameters is difficult [27].

The electrochemical characterization of the electrodes and MEAs in PEMFCs has been widely studied. Electrochemical impedance spectroscopy (EIS) is increasingly utilized by researchers to study the performance of proton exchange membrane (PEM) fuel cell [21,28,29].

In this study, the manufacture of the electrode and MEAs is performed with varying quantities of catalyst content on the activated carbon. The electrode is manufactured using the spraying method on a GDL. This study focuses on attempts to characterize the electrode and test the MEA performance under various conditions.

2.   Material and methods

The materials used to manufacture the electrodes were P75T CPS carbon paper (Ballard, USA) as a gas diffusion media (GDM) or macroporous layer (first layer), carbon black-Vulcan XC72 (Cabot, USA) as carbon substrate, PTFE solution (60 wt%, DuPont, USA), Nafion Solution (5 wt%, DuPont, USA), Pt/C (20 wt%, PMC, USA), MPL and CL. The MPL of the second layer electrode was made by ultrasonicating carbon black – Vulcan XC72 (3.0 mg cm^{-2}), isopropyl alcohol (IPA) and ammonium bicarbonate (50 wt%) for 17 min, then incorporating 30 wt% PTFE for carbon mass and sonicating for an additional 5 min. The resulting ink was sprayed uniformly onto the carbon paper and then placed in a furnace at 350 °C for 3 h. The third layer of the electrode was a CL with a platinum content at the anode of 0.1, 0.3, 0.5, 0.7 and 0.9 mg cm^{-2} with a constant platinum content at the cathode of 0.7 mg cm^{-2}. The CL consists of three layers: The first layer was made by mixing half of the total Pt/C content with 20 wt% PTFE and IPA, spraying the mixture onto the GDL and sintering at 350 °C for 3 h. The second catalyst layer consists of remaining Pt/C, IPA and 17.5 wt% Nafion for the anode with 15 wt% for the cathode. The third layer consists of 17.5 wt% Nafion and the IPA. The resulting electrodes were analyzed and characterized using SEM–EDX (Zeiss Supra-55 VP).

The cathode and anode with an active area of 25 cm^2 were combined using nafion membranes 212 (NR212) and nafion membranes 112 (N112) for comparison (DuPont, USA), to create the MEA. The performance of the MEAs was tested in a multiple-serpentine flow channel pattern cell using GasHub fuel cell station both with and without back pressure and varying the cell temperature, catalyst content and membranes. Electrochemical characterization of the MEAs was performed using electrochemical impedance spectroscopy (EIS) with a Metrohm Autolab PGSTAT128N (Netherlands). Pure hydrogen gas was supplied to the anode at a flow rate of
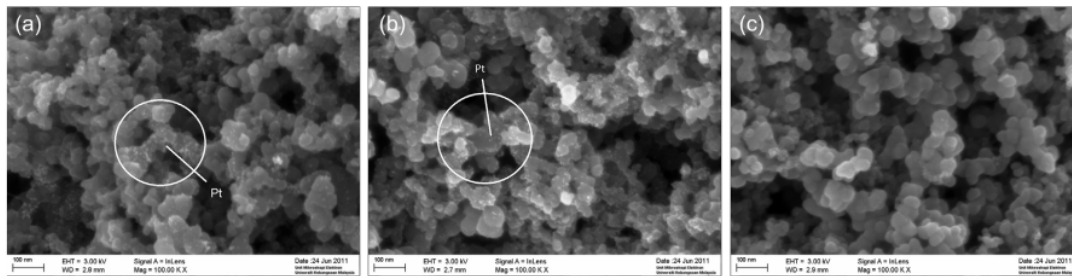
Fig. 1 — The sample SEM image of the electrode (a) anode 0.5 mg cm^{-2} (b) cathode 0.7 mg cm^{-2} (c) GDL.

0.9 L min^{-1}, and purified air was supplied to the cathode at 1.8 L min^{-1} with 100% relative humidity.

3. Results and discussion

3.1. EDX–SEM characterization of the electrodes

The surface and cross-section electrodes with varying platinum catalyst content were analyzed using FE-SEM with 100,000× magnification to observe the distribution of the platinum particles on the surface and in cross-sectional slices of the electrodes. An SEM image of the CL at the same magnification was used to ensure that the small granules on the electrode contained platinum.

Fig. 1 shows small grains of platinum dispersed on the carbon surfaces of the CL with no observable platinum grains on the GDL. The image of the surface structure that indicates the existence of Pt on the CL was obtained using EDX mapping.

The cross section of the SEM image indicates that increasing the catalyst load increases thickness of the electrode (Fig. 2 and Table 1).

Fig. 2 indicates that the electrode consists of three components: carbon paper as a GDM or macroporous layer, a MPL

and a CL. Table 1 shows that the average thickness of the electrodes increases with increasing catalyst content. An increase in the platinum content causes an increase in the carbon content so that the overall thickness of the electrode increases. A thicker electrode has a higher mass transfer resistance and a lower electrical conductivity.

SEM–EDX results for the electrode with a 0.1 mg cm^{-2} catalyst content (for example) are shown in Fig. 3. Surface mapping indicates that Pt was scattered on the surface of electrode with the Pt content determined by EDX measurements. In addition to Pt, Fluorine (F) from the Nafion was detected. Based on EDX, the Pt content on the electrode was determined to range between 7.5 and 21.5 wt%. The functionality of the catalyst in the CL not only depends on the content of the catalyst but also on the distribution of the catalyst on the CL surface. EDX mapping can be one mechanism to determine the functionality of the catalyst.

3.2. Performance measurement of the MEA

The MEA performance tests include varying the catalyst content, temperature and membranes thickness. The electrolyte membrane used in this study was NR212 with N112 used for comparison.

3.2.1. The effect of Pt catalyst content
The effect of the Pt content on the MEA performance is based on the function of the catalyst in the electrochemical process at the catalyst layer. The reaction of hydrogen and oxygen to produce water and electricity occurs on the active site of the catalyst. Generally, an increase in the catalyst content will
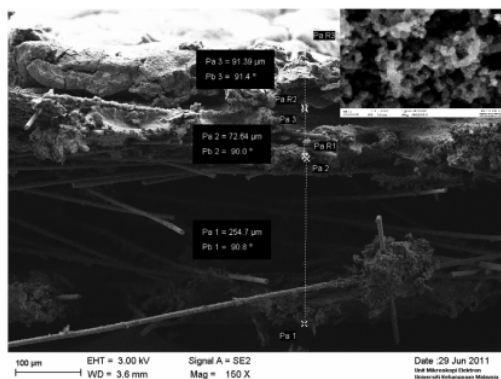

Fig. 2 — Cross-sectional SEM observations of electrode at 0.1 mg cm^{-2} catalyst content.

Table 1 — The electrode thickness data on the various content of the catalyst.

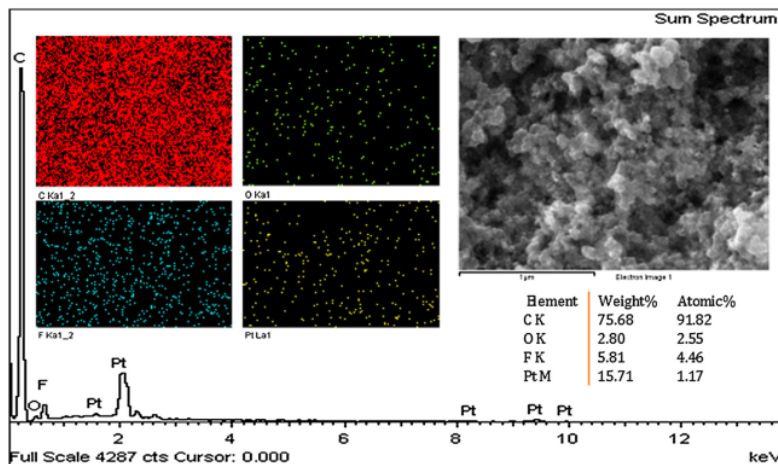| Catalyst content (mg cm^{-2})	Average thickness of electrode (μm)
0.1	329.83
0.3	375.16
0.5	389.07
0.7	437.37
0.9	443.80

Fig. 3 — The result of EDX and mapping electrode with 0.1 mg cm^{-2} Pt catalyst content.

increase the performance of the MEA due to an increase in the number of active sites. However, it depends on the effectiveness of catalyst distribution and the catalyst particle size.

Fig. 4 shows the performance of the MEAs with varying catalyst content. In generally, the performance of MEAs increase with increasing catalyst content, until the optimum active surface area of catalyst is reached. This occurs because the surface area decreases with increasing content of the catalyst [23]. Fig. 5 shows the current density operation points for various Pt contents at a cell voltage value of 0.6 V.

The current density increases with increasing Pt catalyst content up to 0.5 mg cm^{-2} as illustrated in Fig. 5. A catalyst content greater than 0.5 mg cm^{-2} leads to decreasing MEA performance. This decrease is most likely due to a buildup of the catalyst in which the Pt covers itself and causes a reduction in the number of active catalyst sites. Fig. 6 indicates that a distribution of the catalyst with a catalyst content of 0.5 mg cm^{-2} is more homogenous and more even than a catalyst content of 0.7 or 0.9 mg cm^{-2}. Electrochemical characterization of the electrode as illustrated in Fig. 7 confirms that an electrode with a Pt content of 0.5 mg cm^{-2} performs better than one with a Pt content of 0.7 or 0.9 mg cm^{-2} as indicated by the reduced impedance.

The single impedance arc of the MEA as show in Fig. 7 indicates that the electrode process is dominated by the interfacial kinetics of the redox reaction at the electrode, especially the ORR process [21]. The reduced resistance at the high frequency intercept indicates that the total ohmic resistance of the cell (the sum of the contributions from the contact resistance between the components and the ohmic resistance of the cell components) is low.

3.2.2. The effect of temperature and back pressure
The performance of the MEA increases with increasing operational temperature as shown in Fig. 8. The increase in temperature will increase the kinetic energy of the gas (corresponding Arrhenius equation) so that electrochemical reactions become more effective and increase the fraction of
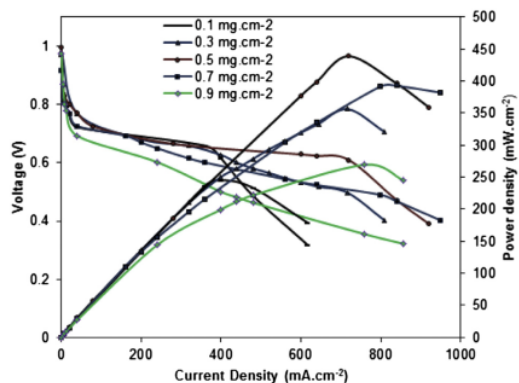

Fig. 4 — Relation of catalyst content to performance of MEA at ambient temperature condition.
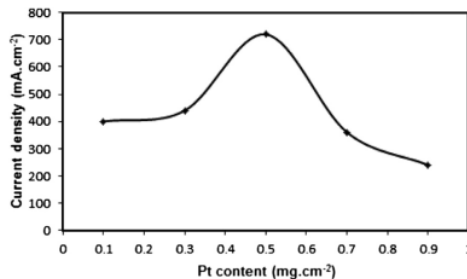

Fig. 5 — The current density operation points for various Pt contents at a cell voltage value of 0.6 V.
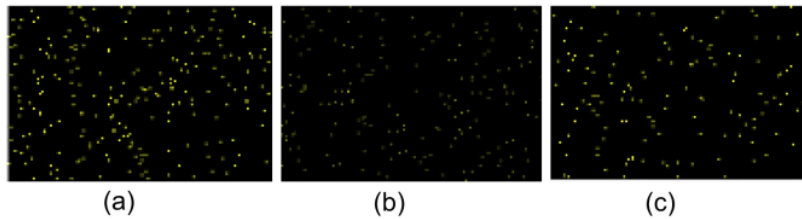
Fig. 6 – The EDX–SEM image of distribution of catalyst at electrode with the catalyst content (a) 0.5 mg cm^{-2} (b) 0.7 mg cm^{-2} (c) 0.9 mg cm^{-2} respectively.
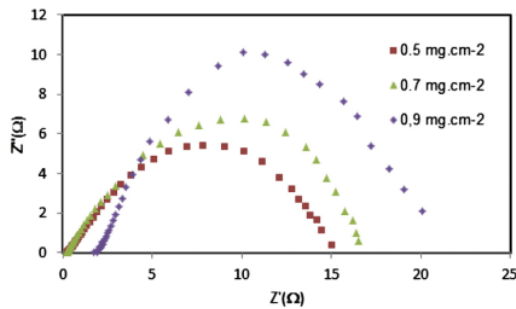

Fig. 7 – Comparison of impedance plots at different Pt contents.

molecules that provide the activation energy required for the reaction (according to the Maxwell–Boltzmann distribution curve). The effect of temperature on the performance of the MEA was also demonstrated by Su et al. [12].

Based on Fig. 9, the performance of the MEA increased sharply when the fuel cell temperature was raised from ambient to 50 °C (with increasing average power density 25.5%) and continued to increase with increasing temperature, although the increase is not significant enough on power density. The increase in the average power density from 50 °C to 60 °C only about 1%. Meanwhile, the increase in power density of 60–70 °C is 0.3%. In addition, based on the Gibbs free energy equation for a reaction with a negative entropy

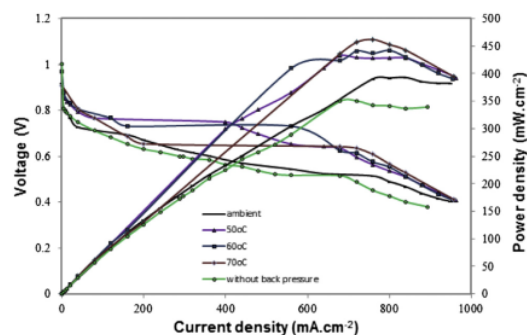change, an increase in temperature reduces the spontaneity of the reaction. The cell temperature and input temperature of the anode and cathode must be balanced [25]. If the input temperature is lower than the temperature of the cell, drying will occur in the cell, and if the input temperature is higher than the cell temperature, flooding will occur. Both circumstances will degrade the performance of the PEMFC.

Figs. 8 and 9 also indicate that the performance of the MEA increased by approximately 25% at 0.6 V with the application of a 10-psi back pressure. Back pressure can increase the permeability of hydrogen and oxygen to the electrode so that more hydrogen and oxygen are available to react. The influence of back pressure can also be observed in Fig. 10. The diameter of the kinetic loop decreases with the addition of back pressure, indicating a reduction in the MEA resistance. In addition, the effect of temperature on the resistance of the MEA was evidenced by a decrease at 50 °C.

3.2.3. Electrolyte membrane effect

The performance of the MEAs using NR212 was improved relative to those using N112 as shown in Fig. 11. The membrane performance is closely related to the level of ionic conductivity which describes the ability of proton migration in the aqueous phase and proton tunneling between adjacent sulfonate groups in narrow pores. Studies on the conductivity of the membranes were carried out by several researchers [30–33]. They have reported that the conductivity of fully hydrated membrane is enhanced with membrane thickness and the operating temperature [30]. In addition, the relationship between the thickness of the membrane with osmotic
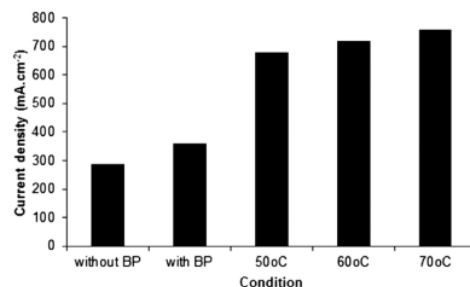

Fig. 8 – The effect of temperature and back pressure to performance of MEA with 0.7 mg cm^{-2} catalyst content.

Fig. 9 – The current density operation points for various conditions at a cell voltage value of 0.6 V.
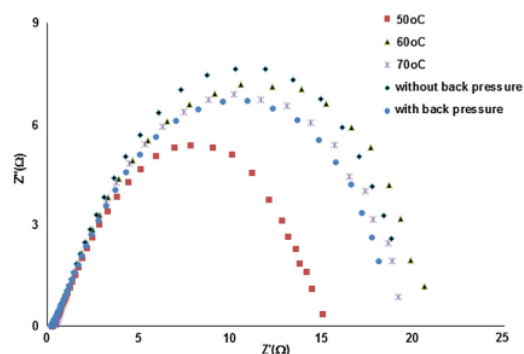
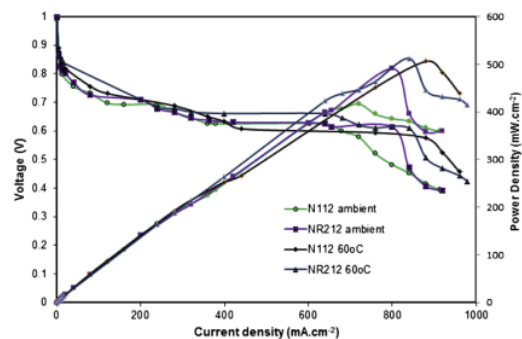Fig. 10 — Comparison of impedance plots at different conditions.

Fig. 11 — The effect of different electrolyte membranes to MEA performance.

drag coefficient (the number of water molecules dragged by each proton from the anode to the cathode) is also enhancing the performance [34]. According to the characteristics of the membrane NR212 and N112 provided by DuPont Fuel Cell USA, acid capacity (Ion Exchange Capacity/IEC) of NR212 (0.95 − 1.01 meq/g) is greater than N112 (0.89 meq/g). The IEC value is related to water uptake properties (WU) of the membranes. The value WU of NR212 and N112 is 50% and 38%, respectively. These values are closely related to the value of proton conductivity of the membrane as has been shown by Watari [35]. Watari shows that the conductivity increases up to an optimum value with increasing water uptake. Meanwhile, Cooper [33] reported that the NR212 has more uniform conductivity than N112, both in-plane direction and through-plane. While for N112, it has greater in-plane conductivity compared to its own Through-Plane conductivity. In general, the conductivity increases with humidity.

4.   Conclusions

The distribution of platinum on the electrode plays an important role for catalysts associated with electrochemical reactions. In addition, the content of the catalyst, the cell temperature, the back pressure and the type of membrane affect the performance of the MEA. MEA performance increases with increasing catalyst content until an optimal catalytic active site is achieved. Likewise, an increase in the cell temperature causes an increase in the kinetic energy of the reaction and the performance of MEA until the optimum temperature conditions are met. Back pressure can improve the performance of the MEA by increasing the quantity of hydrogen and oxygen available to react at the catalyst layer. A Nafion 212 membrane exhibited better performance than the 112 membrane, especially at high temperatures.

Acknowledgments

R E F E R E N C E S

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