

Effect of Hydrogen Flow Rate on MEA Performance with a Three-Catalyst-Layer Pt/C Configuration

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Abstract

An essential component in Proton Exchange Membrane Fuel Cells (PEMFCs) is the Membrane Electrode Assembly (MEA), which facilitates the electrochemical reaction between hydrogen and oxygen to generate electrical energy. This study examines the effect of varying hydrogen gas flow rates on the performance and durability of a Pt/C-based MEA. The MEA used in this research measures 6.5 cm × 30 cm with a catalyst loading of 2 mg/cm². The electrode is constructed in three layers of catalysts to maximize interfacial contact within the catalyst layer. The tested hydrogen flow rates were 100, 200, 300, and 400 ml/min. Performance evaluation was conducted through polarization (I–V) and power (I–P) curve measurements. The results indicated optimal performance at a 200 ml/min flow rate, with a maximum power density of 3.563 mW/cm² and a current density of 10.256 mA/cm². Durability testing was carried out under a constant current of 2 A for 12 hours and showed a voltage drop of 24.35% after 10 hours of operation. Electrochemical characterization using Cyclic Voltammetry (CV), Electrochemical Impedance Spectroscopy (EIS), and Linear Sweep Voltammetry (LSV) yielded an Electrochemical Surface Area (ECSA) of 1.477×10^{-5} m²/g, electrical conductivity of 3.218×10^{-4} S/cm, and an electric charge of 4.2×10^{-6} C.

Keywords: *electrode, three-catalyst-layer, hydrogen flow rate, Pt/C, PEMFC*

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Komponen yang esensial dalam *Proton Exchange Membrane Fuel Cell* (PEMFC) yakni *Membrane Electrode Assembly* (MEA) yang berperan memfasilitasi reaksi elektrokimia antara hidrogen dan oksigen untuk menghasilkan energi listrik. Penelitian ini bertujuan mengkaji pengaruh variasi laju alir gas hidrogen terhadap performa dan durabilitas MEA berbasis katalis Pt/C. MEA yang digunakan berukuran 6,5 cm × 30 cm dengan beban katalis 2 mg/cm². Elektroda disusun dalam tiga lapisan untuk memaksimalkan kontak antarmuka pada lapisan katalis. Variasi laju alir hidrogen yang diuji meliputi 100, 200, 300, dan 400 ml/menit. Evaluasi kinerja dilakukan melalui pengukuran kurva polaritas (I–V) dan kurva daya (I–P). Hasil menunjukkan performa optimum pada laju alir 200 ml/menit, dengan densitas daya maksimum sebesar 3,563 mW/cm² dan densitas arus sebesar 10,256 mA/cm². Uji ketahanan dilakukan pada arus konstan 2 A selama 12 jam dan menunjukkan penurunan tegangan sebesar 24,35% setelah 10 jam operasi. Karakterisasi elektrokimia menggunakan teknik *Cyclic Voltammetry* (CV), *Electrochemical Impedance Spectroscopy* (EIS), dan *Linear Sweep Voltammetry*

(LSV) menunjukkan nilai *Electrochemical Surface Area* (ECSA) sebesar $1,477 \times 10^{-5} \text{ m}^2/\text{g}$, konduktivitas listrik $3,218 \times 10^{-4} \text{ S/cm}$, dan muatan listrik sebesar $4,2 \times 10^{-6} \text{ C}$.

Kata Kunci: elektroda, tiga lapisan katalis, laju alir hidrogen, Pt/C, PEMFC

INTRODUCTION

Hydrogen is a renewable and environmentally friendly energy source, as it produces water as a by-product and does not emit pollutants into the environment [1]. Recognized as the cleanest energy source in the world, hydrogen is abundantly available, highly flexible, and holds great potential to support sustainable development agendas [2]. Hydrogen can be utilized as a fuel in fuel cells through an electrochemical process that directly converts its chemical energy into electrical energy through reduction-oxidation reactions [3].

One of the most widely used and developed types of fuel cells is PEMFC [4,5]. PEMFC is an energy conversion device that uses an acid-based electrolyte membrane to facilitate the transfer of protons from the anode to the cathode, while electrons flow through an external circuit to generate an electric current [6]. The main advantages of PEMFC include high power density, good energy conversion efficiency, relatively low operating temperatures, zero emissions during operation, compact structure, and environmental friendliness. In addition, PEMFCs offer fast start-up times and operate silently [7].

The Membrane Electrode Assembly (MEA) is a critical component in PEMFCs, acting as the heart of the electrochemical system within the fuel cell [8]. It plays a central role in facilitating electrochemical reactions by enabling proton transport and electron separation, while also serving as a barrier to prevent the mixing of reactant gases at the anode and cathode [9]. The efficiency and operational stability of a PEMFC system are largely dependent on the performance of the MEA. A key factor affecting MEA performance is the uniform distribution of gases and water within the Gas Diffusion Layer (GDL) and across the catalyst layer [10].

GDL has a porous structure located between the catalyst layer and the gas flow channels, serving to ensure uniform distribution of reactant gases to the catalyst surface, while also providing mechanical support and enabling the removal of water produced during the electrochemical reaction [11]. During PEMFC operation, electrochemical reactions at the catalyst layers of the electrodes generate electrical energy while producing water as a by-product [12]. A key characteristic of an effective fuel cell catalyst is high electrocatalytic activity, particularly in the

oxygen reduction reaction (ORR), as seen in platinum-based (Pt) catalysts [8]. Carbon-supported platinum (Pt/C) remains the most commonly used commercial catalyst in PEMFC applications due to its excellent performance and stability [13,14].

The presence of a catalyst layer on the electrode requires meticulous attention, as improper application may result in catalyst degradation. In this study, electrode formation with catalyst application on the GDL was carried out using three catalyst layers to ensure homogeneous catalyst distribution and to maximize interlayer contact for optimal electrochemical performance. Furthermore, the effect of hydrogen gas flow rate on the performance of the MEA was evaluated. The hydrogen supply rate can either enhance or hinder MEA performance, depending on the balance between fuel availability and the requirements of the electrochemical reaction [5]. Consequently, performance tests were conducted to determine the optimal operating conditions, and MEA durability was assessed by evaluating the optimal current derived from variations in hydrogen flow rate to ensure long-term stability.

MATERIALS AND METHODS

Materials

The materials used in this study include 2-propanol ($\text{C}_3\text{H}_8\text{O}$) (Merck), ammonium bicarbonate (NH_4HCO_3) (Merck), P75T CVSP carbon paper, deionized water, Vulcan XC-72R carbon, Pt/C 40% catalyst (Fuel Cell Store), laminating film, Nafion™ 35% ionomer solution (Dupont), 60% polytetrafluoroethylene (PTFE) emulsion D-2020, and Nafion™ 212 membrane.

Methods

Fabrication of the three-catalyst-layer electrode

The catalyst loading of Pt/C used was 2 mg/cm^2 . The catalyst ink mixture for the first layer catalyst consisted of 50 wt.% Pt/C catalyst, PTFE, and 2-propanol. The mixture was homogenized using an ultrasonic homogenizer for 10 minutes, then transferred to a spray gun and sprayed onto the GDL measuring $13 \text{ cm} \times 30 \text{ cm}$. The second catalyst layer of the electrode consisted of 50 wt.% Pt/C catalyst, 50 wt.% Nafion solution, and 2-propanol. This mixture was also homogenized using an ultrasonic homogenizer for 10 minutes, then sprayed onto the first catalyst layer until the ink was fully applied. The third

catalyst layer consisted of a 50 wt.% Nafion solution and 2-propanol. The mixture was homogenized and sprayed onto the second-layer catalyst to form the three-layer catalyst of the electrode, following the procedure described in [15], while the illustration is presented in **Figure 1**.

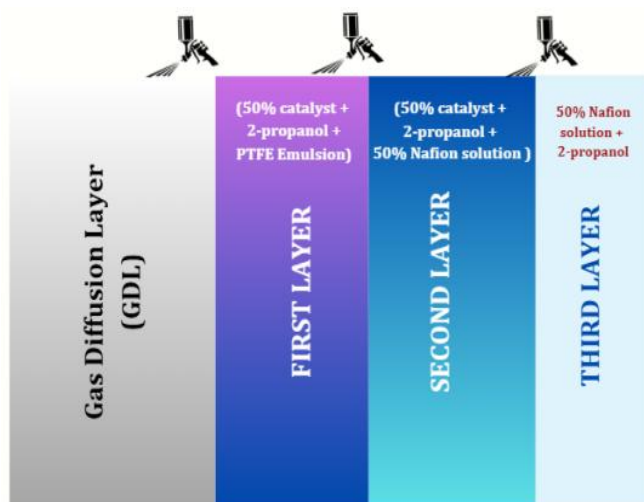


Figure 1. Illustration of the three-layer catalyst of electrode

MEA fabrication

The three-layer catalyst of the electrode was carefully cut into two pieces, each measuring 6.5 cm × 30 cm. A Nafion 212 membrane was positioned between the electrodes on both the anode and cathode sides. The assembly was subsequently covered with laminating film. The electrodes were then enclosed in aluminum foil on both sides, clamped using aluminum plates, and placed at the center of a hot press. The system was heated to 135 °C for 3 minutes under a pressure of 2000 psi.

Performance and durability test of MEA in PEMFCs

Initial testing was performed by measuring the OCV (Open Circuit Voltage), where the voltage was recorded without load at varying hydrogen flow rates of 100, 200, 300, and 400 ml/min until a stable voltage was reached. The performance of the MEA in a single PEMFC was tested under different hydrogen flow rates supplied to the anode, while oxygen was supplied to the cathode using a blower. Measurements were conducted using the Smart-2 Fuel Cell Test Station from WonATech, operated via the FCT Server and WFTS programs. The voltage was recorded for each current increment until the MEA could no longer maintain the voltage. The performance was determined by polarization curves (I-V and I-P).

The durability of the MEA in a single PEMFC stack was measured based on the optimal results from the hydrogen flow rate variation tests using the Smart-

2 Fuel Cell Test Station from WonATech. The MEA, which demonstrated optimal performance under varying hydrogen flow rates, was then tested for durability over time by operating it at the optimal current density load. Durability measurements were taken based on the voltage-time relationship during a 12-hour test period.

Data analysis

CV analysis is employed to determine the Electrochemical Surface Area (ECSA), which allows the calculation of the active surface area of the catalyst in the MEA, as described by Equation (1) [16].

$$ECSA = \frac{Q}{210 \mu C/cm^2 \times m_{catalyst}} \quad (1)$$

In this method, Q represents the average charge obtained from the hydrogen adsorption/desorption region in the cyclic voltammetry (CV) curve. A value of 210 $\mu C/cm^2$ is commonly used as the charge required for monolayer hydrogen adsorption on platinum (Pt) surfaces, serving as a standard for ECSA calculations. Catalyst loading (C/m^2) refers to the mass of catalyst deposited per unit area of the MEA and plays a key role in evaluating the available active sites.

Meanwhile, electrochemical Impedance Spectroscopy (EIS) is employed to obtain Nyquist plots, which are then fitted to extract key parameters such as the polarization resistance (R_p) and the solution resistance (R_s). These values are essential for evaluating the electrical conductivity of the electrode, which can be calculated using the following Equation (2).

$$\sigma = \frac{1}{Z_R} \times \frac{l}{A} \quad (2)$$

where σ represents the electrical conductivity (S/cm), l is the thickness of the sample (cm), R is the total resistance ($R_p + R_s$) (ohm), and A is the cross-sectional area of the electrode (cm^2) [17].

The electrochemical properties of the MEA are determined using the Linear Sweep Voltammetry (LSV) method, where the electric charge (Q) is calculated using Equation 3.

$$\begin{aligned} V &= vt \\ dt &= \frac{dV}{v} \\ I &= \frac{dQ}{dt} \\ \frac{I}{v} &= \frac{dQ}{dV} \\ Q &= \frac{I \times \Delta V}{v} \end{aligned} \quad (3)$$

where Q is the electric charge (C), I is the peak current (A), ΔV is the voltage change (V), and v is the scan rate (V/s) [18].

RESULTS AND DISCUSSION

Electrode morphology

GDL on the electrode serves as a mechanical support for the MEA and facilitates the transport of substances involved in the reaction, such as reactant gases and water products, through interconnected pores. The morphology of the GDL can be seen in **Figure 2**.

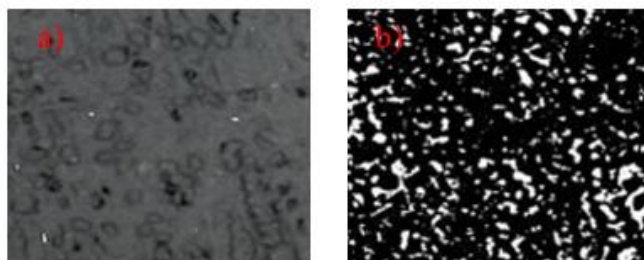


Figure 2. GDL surface images obtained using a) digital microscope and (b) ImageJ software.

Figure 2 (a) illustrates the GDL morphology, showing carbon pores coated with carbon ink, which forms the Micro Porous Layer (MPL). The GDL morphology was then processed using the thresholding method in ImageJ, resulting in the black-and-white image shown in **Figure 2 (b)**. In this image, the black areas represent the pores, while the white areas indicate the distribution of solid particles. The pores are evenly distributed across the GDL surface, albeit with varying sizes, reflecting a high level of porosity. The formation of the pores is influenced by the inclusion of ammonium bicarbonate in the preparation of the carbon ink [19].

The pore distribution in GDL and the MPL, as shown in **Figures 2(a)** and **2(b)**, plays a crucial role in supporting the transport of reactant gases and the removal of water products in the fuel cell. The evenly distributed pores, although varying in size, indicate a high porosity level, which directly impacts the diffusion effectiveness and overall performance of the fuel cell [20].

The multilayer design in the fabrication of the three-layer catalyst of the electrode facilitates a more homogeneous distribution of active materials, enhances the contact between the catalyst and ionomer, and enables more precise control over porosity. This stratified structure also aids in minimizing mass transport resistance and optimizing charge transfer efficiency [15]. Collectively, these improvements contribute to the enhancement of output power and

operational stability of the MEA in PEMFC applications. The morphology of the three-layer catalyst of the electrode is illustrated in **Figure 3**.

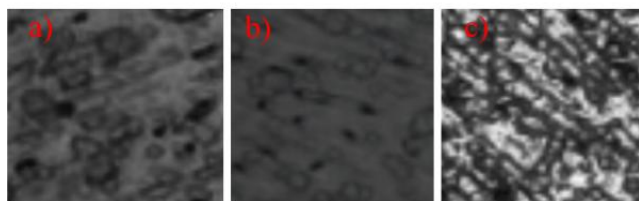


Figure 3. Pt/C electrode with three-layer catalyst (a) first layer; (b) second layer; (c) third layer

The morphology of the three-layer electrode, as depicted in **Figure 3**, illustrates a relatively uniform pore distribution on the surface of the GDL. In **Figures 3 (a)** and **(b)**, the pores on the electrode surface remain discernible. In contrast, **Figure 3 (c)** demonstrates the even sealing of the electrode's pores with Nafion. Nafion, a perfluorosulfonic acid-based ionomer (PFSA), exhibits proton conductivity and is frequently incorporated into the catalyst and support mixture to enhance proton conductivity while facilitating the adhesion of the catalyst layer. The porosity and pore distribution within the electrode are of paramount importance in the diffusion processes and the transport of reactants to the catalyst layer, significantly influencing the overall electrochemical performance [21].

Membrane electrode assembly (MEA)

The MEA is fabricated by sandwiching a Nafion 212 membrane between two electrodes, the cathode and the anode, each measuring 6.5 cm × 30 cm. The assembly process involves hot-pressing, which applies substantial pressure to fuse the layers of the MEA components, electrodes, and Nafion 212 membrane—thereby creating a robust interface between the three constituents. This strong bond is essential for ensuring optimal contact between the catalyst on the electrodes and the proton-conducting Nafion membrane, thereby facilitating efficient electrochemical reactions. The resulting MEA, with an area of 6.5 cm × 30 cm, is depicted in **Figure 4**.



Figure 4. The fabricated of MEA

Figure 4 illustrates that the electrodes and Nafion membrane are tightly bonded without any visible air bubbles or voids around the electrodes. This intimate adhesion is crucial for ensuring that there is no impedance or disruption in the electrochemical process. The absence of voids enhances the efficiency of proton transfer from the Nafion membrane to the catalyst on the electrode, as well as the effective transport of reactants and products through the electrode layers. Such a well-aligned structure ensures maximal contact between the catalytic sites and the proton-conducting medium, thereby facilitating optimal electrochemical performance in the MEA.

Electrochemical analysis

Electrochemical analysis of the MEA was carried out using CV, EIS, and LSV. The CV measurements were conducted with a scan rate of 50 mV/s over a potential window ranging from -0.39 V to 1.1 V, resulting in the voltammogram shown in **Figure 5a**. From the voltammogram, the Electrochemical Surface Area (ECSA) was calculated based on the double-layer capacitance, yielding a value of 1.477×10^{-5} m²/g. A high ECSA value indicates a greater number of electrochemically active sites on the electrode surface and reflects low charge transfer resistance, both of which are essential for enhancing the proton conductivity and overall electrochemical performance of the MEA.

The EIS analysis produced a Nyquist plot, as illustrated in **Figure 5b**, which displays a characteristic semicircular arc indicative of the interplay between the real and imaginary components of the impedance spectrum [14]. The diameter of the semicircle corresponds to the total cell resistance, which was determined to be $1.5937 \times 10^5 \Omega$. Based on this resistance value and the known geometric parameters of the cell, the calculated electrical conductivity was 3.218×10^{-4} S/cm. This relatively low conductivity is attributed to the intrinsic electronically insulating nature of the proton exchange membrane, which is designed to selectively conduct protons while blocking electron transport. This selective property is essential for PEMFC operation, ensuring that electrons are directed through an external circuit to generate useful electrical power, thereby maintaining electrochemical efficiency and system functionality.

In addition, LSV was conducted under conditions similar to CV, but with a unidirectional potential sweep. The resulting voltammogram, presented in **Figure 5c**, demonstrates the current response to varying applied potential, allowing for the evaluation of redox activity within the electrode interface. The calculated charge transfer (Q) derived from the area under the curve was 4.2×10^{-6} C, indicating the electrochemical reactivity and redox dynamics of the active species present in the MEA system.

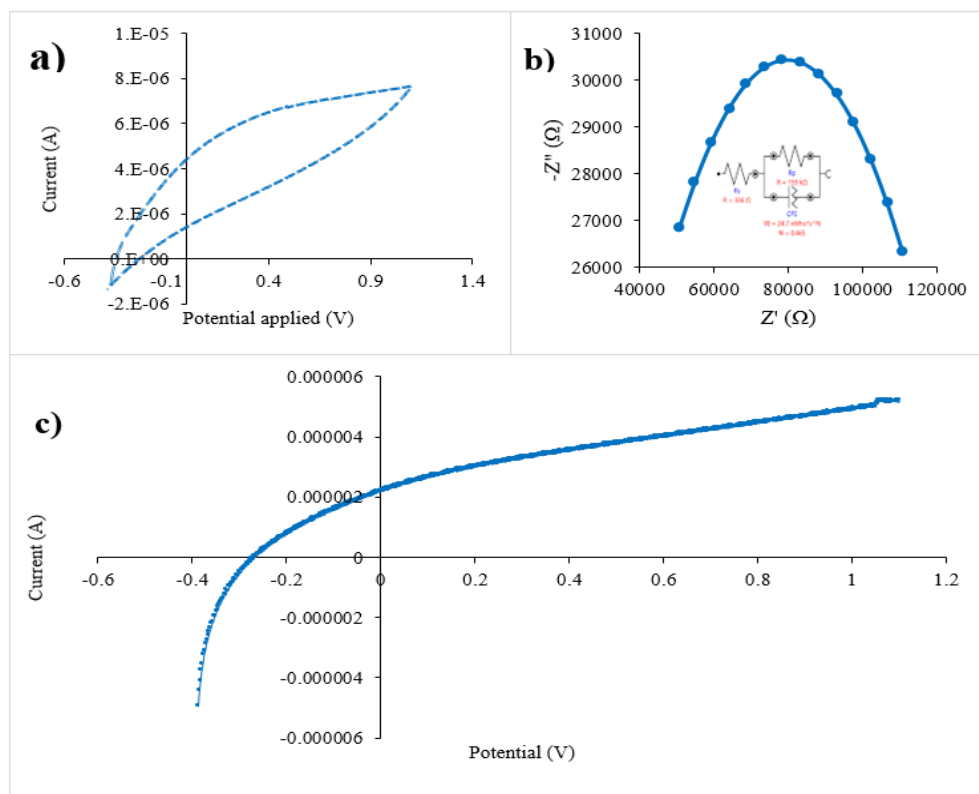


Figure 5. a) CV voltammogram, b) Nyquist curve, and c) LSV curve of the MEA with Pt/C Catalyst

Measurement of OCV with variations in hydrogen flow rate

The initial performance evaluation of the MEA was carried out by determining the OCV, which is the voltage measured without any load applied. Based on the tests conducted, the OCV values can be seen in Figure 6.

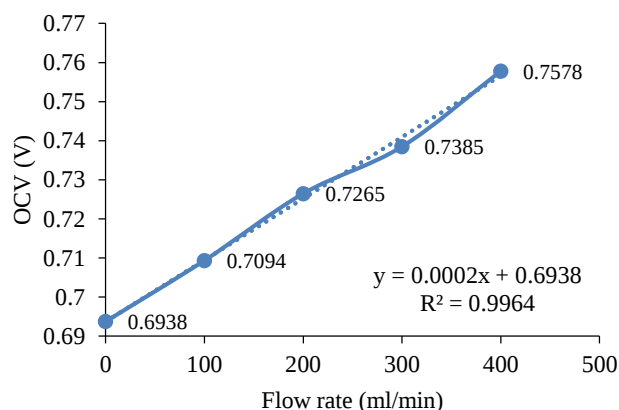


Figure 6. The OCV of MEA with variation of hydrogen flow rate using Pt/C catalyst in a PEMFC

Figure 6 shows an increase in OCV with the rise in hydrogen flow rate, with the highest OCV value of 0.758 V obtained at a flow rate of 400 ml/min. A higher hydrogen supply results in a greater number of dissociated protons and electrons, enabling more optimal electrochemical reactions at a flow rate of 400 ml/min, thereby increasing the OCV.

Based on the obtained OCV values, a linear relationship between hydrogen flow rate and OCV can be established. Figure 6 illustrates this linear relationship, with an intercept value of 0.694 and a slope of 0.0002. Additionally, the coefficient of determination (R^2) is 0.9929, indicating a very strong linear correlation. Therefore, the model equation representing the relationship between OCV and hydrogen flow rate can be written as:

$$\text{OCV} = 2 \times 10^{-4} v + 0.694$$

This equation indicates that when no hydrogen is supplied (i.e. hydrogen flow), the OCV value is 0.694 V.

MEA Performance with variations in hydrogen flow rate

The performance of the MEA using a Pt/C catalyst in a PEMFC was evaluated at hydrogen flow rates of 100, 200, 300, and 400 ml/min. The MEA performance was assessed based on I-V and I-P curves, as shown in Figures 7a and 7b.

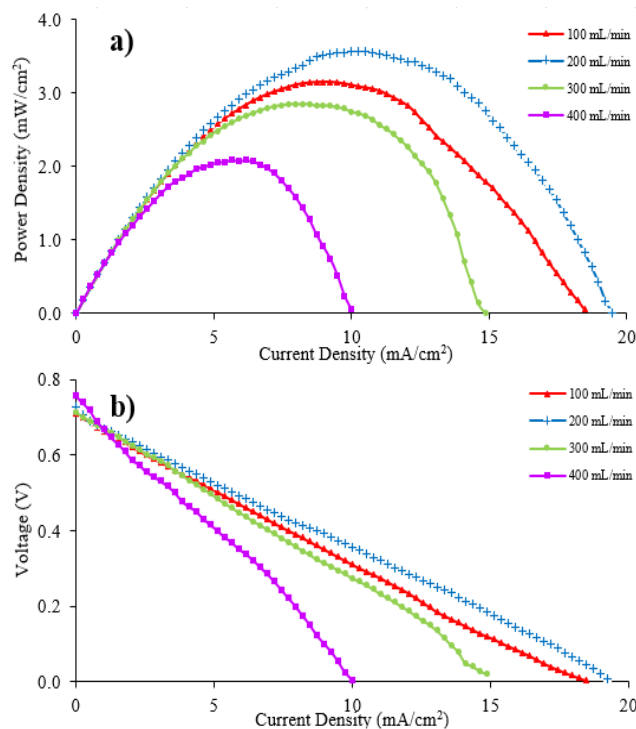


Figure 7. Polarization curves of the MEA at varying flow rates a) I-V dan b) I-P

Figure 7 illustrates the effect of hydrogen flow rate on the MEA performance. The optimal performance is observed at a 200 ml/min flow rate. At a lower flow rate of 100 ml/min, the performance is reduced due to insufficient hydrogen supply. Conversely, at higher flow rates of 300 and 400 ml/min, performance declines because the excessive hydrogen supply cannot be fully utilized by the MEA, leading to suboptimal reactions. Figure 7 illustrates the effect of hydrogen flow rate on MEA performance. Optimal performance is achieved at a flow rate of 200 ml/min. At a lower flow rate of 100 ml/min, performance decreases due to insufficient hydrogen supply. Conversely, at higher flow rates of 300 and 400 ml/min, performance declines because the excessive hydrogen supply cannot be fully utilized by the MEA, leading to suboptimal reactions.

Excessive hydrogen supply can lead to hydrogen crossover due to high pressure, where hydrogen moves from the anode to the cathode through the membrane without dissociating into protons. This direct reaction between hydrogen and oxygen is exothermic and can generate hotspots on the membrane, potentially causing small holes (pinholes). This condition exacerbates crossover phenomena, accelerates membrane degradation, reduces fuel cell efficiency, and threatens the safety and lifespan of the fuel cell [23].

MEA durability

Durability testing is conducted to analyze the performance degradation of the MEA over time. After evaluating the MEA performance at varying hydrogen flow rates, the optimal flow rate and current were determined for the durability test. Based on the MEA performance results, the durability test was performed at a hydrogen flow rate of 200 ml/min and a 2 A load for 12 hours. The measurement results of the MEA durability test are shown in **Figure 8**.

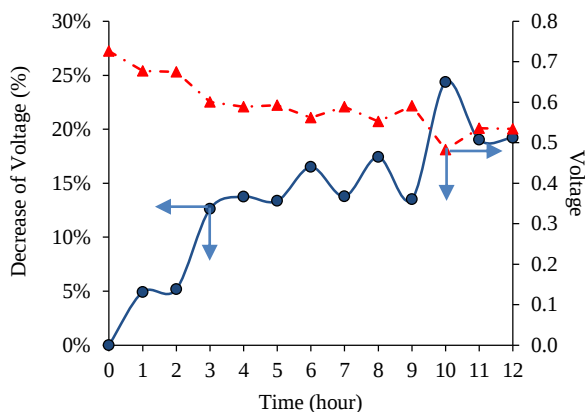


Figure 8. Durability Decrease of MEA with Pt/C Catalyst in PEMFC

The percentage decrease in MEA durability is determined by comparing the voltage at a specific time to the OCV value. **Figure 8** shows that during the first 1 and 2 hours, the voltage drop percentage was 4.92% and 5.12%, respectively. Between 3 to 9 hours, the voltage decreased by an average of approximately 15%. After 10 hours, the voltage drop reached 24.35%. This indicates that the MEA with Pt/C catalyst in PEMFC exhibits good durability, as after 10 hours of testing, the voltage drop is only 24.35%.

CONCLUSION

The Pt/C catalyst-based MEA with a three-layer electrode configuration shows the best performance at a hydrogen flow rate of 200 ml/min, achieving a maximum power density of 3.563 mW/cm² and a current density of 10.256 mA/cm². The layered electrode structure provides a more efficient gas and ion transport path, enhancing catalyst utilization. Durability testing shows a voltage drop of 24.35% after 10 hours of operation at a constant current of 2 A, indicating adequate operational stability. Electrochemical characterization results show values for ECSA, conductivity, and charge storage that support the overall performance and lifespan of the MEA.

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