

Experimental Study of Oxidant Effect on Lifetime of PEM Fuel Cell

M. M. Hassani^a, S.M. Rahgoshay^{b,1}, M. Rahimi-Esbo^{c,1}, K. Dadashi Firouzjaei^d

^a MSc, School of Mechanical Engineering, Babol "Noshiravani" University of Technology, Babol, Iran.

^b Researcher, PhD, Malek Ashtar University of Technology, Tehran, Iran.

^c Assistant professor, Malek Ashtar University of Technology, Tehran, Iran.

^d Researcher, MSc, Malek Ashtar University of Technology, Tehran, Iran

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Abstract

In recent decades, fuel cells have been widely used in energy generation. In a PEM-FC, considering the specific application, two types of oxidants are used. Durability tests, which are highly costly products, are of crucial importance in evaluating the lifetime of fuel cells. The purpose of the present paper is to investigate the performance of a fuel cell by changing the type of oxidant from air to pure oxygen. Because of the presence of impurities in the air oxidant, a cell with air oxidant is more sensitive to operating conditions than one with pure oxygen. In this experiment, a single fuel cell was assembled and used for testing. The lifetime test was carried out in constant current, and the voltage decay rate was reported. Effects of various parameters, like air stoichiometry, Dew point Temperature, and Pressure, have been investigated. Increasing the stoichiometry of the oxidant to 3 greatly increased the voltage of the fuel cell, but no significant increase in the fuel cell voltage was observed in stoichiometries above this value. A comparison of inlet gas temperatures demonstrated that the fuel cell had the best performance at 75 °C, but due to the fluctuation of the output voltage at this temperature, the temperature was decreased to 65 °C. Finally, upon performing durability test with pure oxygen for 9 hours and comparing the results with those of air oxidant, the possibility of using a fuel cell with two different oxidants has been confirmed.

Nomenclature

BOL	Beginning Of Life
C	Concentration of O ₂ in air
DPT.Si.A	Dew point temp. Single cell inlet anode
DPT.Si.C	Dew point temp. Single cell inlet cathode
Eload	Electron load bank
FM	Flow meter

FPS	Fuel processing (sub-)system
HEX	Heat exchanger
HUM	Humidifier
Idens	Current density
MFC	Mass flow controller
OPS	Oxidant processing (sub-)system
p.Si.A	Pressure single cell inlet anode
p.Si.C	Pressure single cell inlet cathode
Q	Volumetric flow rate
St	Stoichiometry

¹Corresponding author: S.M. Rahgoshay; rahgoshay@mut.ac.ir, majid.rahgoshay@gmail.com

M. Rahimi-Esbo; mrahimi@mut.ac.ir

1. Introduction

In recent decades, fuel cells have been widely used in energy generation. A fuel cell is a device that directly converts chemical energy into electrical energy [1, 2]. Among the various types of fuel cells, the PEMFC has been regarded as a suitable alternative to internal combustion engines due to its high efficiency (43-58%), low operating temperature, and low radiation [3, 4]. In a PEMFC, two types of oxidants, air or pure oxygen, are used according to the specific application. For example, pure oxygen is usually used as the oxidant in mines due to the lack of open air and higher oxygen density in comparison to just air.

However, in some devices, such as hydrogen vehicles, air is chosen as the oxidant due to its accessibility. Durability and lifetime play a vital role in fuel cells since their power decreases over time due to the gradual degradation of their components. Researchers usually perform a long-life parameter test on the fuel cell to determine the rate of performance decay. The lifetime required for transportation application and stationary application is 8,000 and 100,000 hours, respectively [5]. Membrane drying typically occurs on the anode side. On the cathode side, the membrane is in contact with water as a product of the oxygen reduction reaction as well as the moisture content of the reactant gas. However, the main reason for membrane drying is poor water management resulting in decreases in membrane conductivity and increases in ohmic losses. Humidification can improve temporary voltage loss. However, the continued operation of a dry cell over a long time can cause irreversible damage to the membrane. Le Canut et al. [6] performed an experimental study on a fuel cell for 12 minutes in dry conditions. At the beginning of this period, the fuel cell voltage was 0.8v but eventually decreased to 0.75V. After 15 to 20 minutes of humidification, the cell voltage regained its initial level.

Membrane degradation is one of the most important factors in reducing fuel cell lifetime. The chemical stability of the membrane is a key factor in enhanc-

ing the durability of a fuel cell. S.-Y. Ahn et al. [7] assembled a 40 cell stack with air oxidant and 200 cm² active surface area and investigated its lifetime for 1800 hours. At atmospheric pressure and a temperature of 75 °C, the average power of each cell was 0.29 W/cm². After performing various analyses, they found that the catalytic degradation and contamination of the MEA lowered the performance of the fuel cell stack. Dubau et al. [8] tested a 110-cell stack for 12860 hours and also analyzed the MEA both physically and chemically. In another study, Martin et al. [9] investigated several MEAs with a Pt/C catalyst at various Pt loadings. They assembled each of them on a single cell and tested it with dry hydrogen and dry air. They also considered different temperatures and stoichiometric conditions at a constant current density of 200 A/cm². They did not observe any alteration in fuel cell performance up to 1000 hours.

Operating a fuel cell under higher temperatures (>100°C) has several advantages [10]. For example, it enhances the electrochemical kinetics of the fuel cell, which results in higher performance of the cell. Moreover, water management and cooling will also improve due to the temperature difference between the cell and the cooling fluid. The waste heat can be recovered, and CO-poisoning is reduced so that lower quality and purity hydrogen gas can be used [10, 11]. Despite the advantages of operating a fuel cell at a higher temperature, elevated temperatures accelerate the degradation of fuel cell components and adversely affect its long-term efficiency and durability [10-13]. Nandjou et al. [14] performed a durability test on a fuel cell in order to investigate the instantaneous effect of temperature on the degradation mechanism in a fuel cell. They compared the temperature of the experimental results with the numerical results. They also reported a voltage decay of 27 μVh⁻¹ and 18 μVh⁻¹ for the automotive and stationary tests, respectively. Vichard et al. [15] performed a long-term durability test on a commercial 1 kW PEMFC for more than 5000 hr under specific operating conditions to reproducing driving cycles. Their results showed that a low ambient temperature results in better humidification of gases and re-

duces the degradation rate significantly. Yang et al. [16] verified voltage cut-off and lean air operation to increase the durability of the fuel cell stack. Their study confirmed that employing both voltage cut-off and lean air supply improved performance durability threefold. Thomas et al. [17] developed an operational strategy to enhance the lifetime of high-temperature PEMFCs. They reported that the current cycling between $0.2 \text{ A}\cdot\text{cm}^{-2}$ and $0.8 \text{ A}\cdot\text{cm}^{-2}$ with a relaxation time of 2 min leads to a lower degradation rate compared to constant load operation at a current density of $0.55 \text{ A}\cdot\text{cm}^{-2}$.

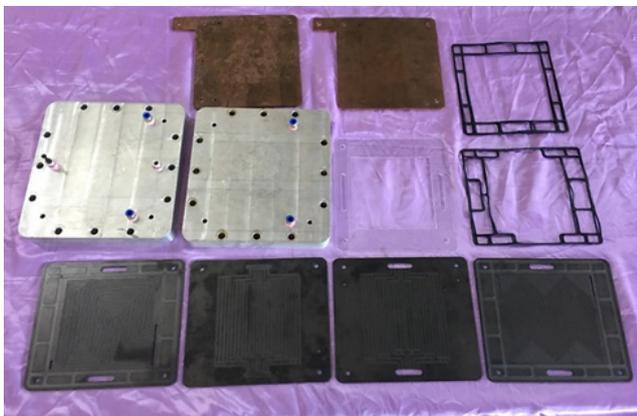
Although many comprehensive studies have been carried out on fuel cells using one oxidant, this paper investigated the degradation rate of the fuel cell performance based on the results obtained by operating the fuel cell with both pure oxygen and air. The possibility of operating a fuel cell using both oxidants for different conditions and applications was also taken into consideration. In other words, the oxidant

type of the cell can be switched to conform to a target application in specific conditions. As a consequence, the cost of constructing a new fuel cell for a specific application would be greatly reduced. Also, the effects of dew point temperature, pressure, and stoichiometry on the performance of this fuel cell have been investigated.

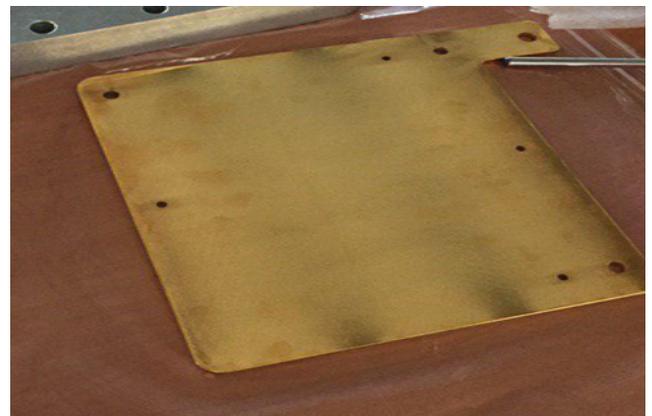
2. Experimental process

2.1. Single cell fuel cell component

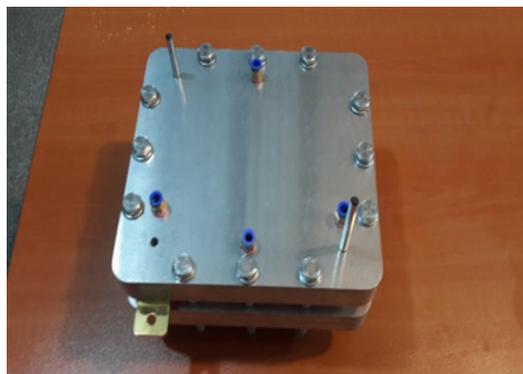
In this experiment, a single fuel cell with an active surface area of 225 cm^2 and copper plates coated with a gold layer, to reduce the contact resistance and enhance the conductivity, were employed. The manufactured components and the assembled PEMFC are shown in Fig. 1.



a)



b)



c)

Fig. 1. Single cell PEMFC with a) manufactured components, b) current collector plate with gold coated, and c) assembled PEMFC.

The polar plates are responsible for adequate and uniform distribution of the reactants, management of generated heat and cooling, collection of current, and the mechanical stability of the fuel cell. A reduction serpentine [18] was used on the anode side of this fuel cell, and a conventional multi-path serpentine flow field was used on the cathode side. In order to prevent leakage of cooling water and gas, the seal is placed on the edges of the flow fields. The multi-layer membrane (MEA) is located between the two gas diffusion layers (like a sandwich), and is considered as the heart of the fuel cell. The MEA used in

this experiment was a PaxiTech with the Nafion 212 membrane. The specifications of the MEA are listed in Table 1. A EPDM seal is used to seal the fuel cell. According to standards [19], the fuel cell should be able to avoid leakage at pressures of two times the operating pressure. In this study, an external leakage test was performed at the pressure of 3 barg, and an internal leakage test was carried out at the pressure of 0.2 barg. In the first case, the pressure drop was less than 0.1 barg for 10 minutes, and in the second case, it was less than 0.01 barg for 10 minutes, which is acceptable according to the standards.



Fig. 2. Leakage test at pressure of 3 barg.

Table 1. Specifications of the PaxiTech MEA.

Component	Unit	Value
GDL (Gas Diffusion layer)		
Area Weight	g/m ²	133
Thickness	μm	251
Compression set at 600 kPa	μm	11
Air permeability through plane [1.22 kPa – 100 ml – 6.42 cm ²]	sec	30
xy air permeability at 600 kPa	μm ²	2.1
Resistivity through plane at 600 kPa (2-point)	mohm.cm ²	10.2
Resistivity in plane (4-point)	ohm	0.8
Catalyst		
Metal concentration	% weight	60-70
Carbon specific area	m ² /g	800
Pt loading	mg/cm ²	0.5
Membrane Nafion 212		
Thickness	μm	50.8
Basic weight	g/m ²	100
Specific gravity	Kg/dm ³	0.97
Available acid capacity (min.)	meq/g	0.92
Total acid capacity	meq/g	0.95 to 1.01
Hydrogen cross-over	ml/min.cm ²	<0.010
Conductivity	S/cm	0.1

2.2. Test procedure

We decided to create artificial air to ensure the exact composition of nitrogen and oxygen gases within the air capsule. To do so, two MFCs were used to make the correct combination of oxygen and nitrogen to form artificial air. To achieve the intended volumetric flow rate, 21% of total volumetric flow was applied to the oxygen’s MFC, and 79% to the nitrogen’s MFC. Fig. 5 shows the MFCs used to generate artificial air. Also, the test apparatus and its schematic diagram are presented in Fig. 3 and Fig. 4, respectively.



Fig. 3. Image of the PEMFC test station.

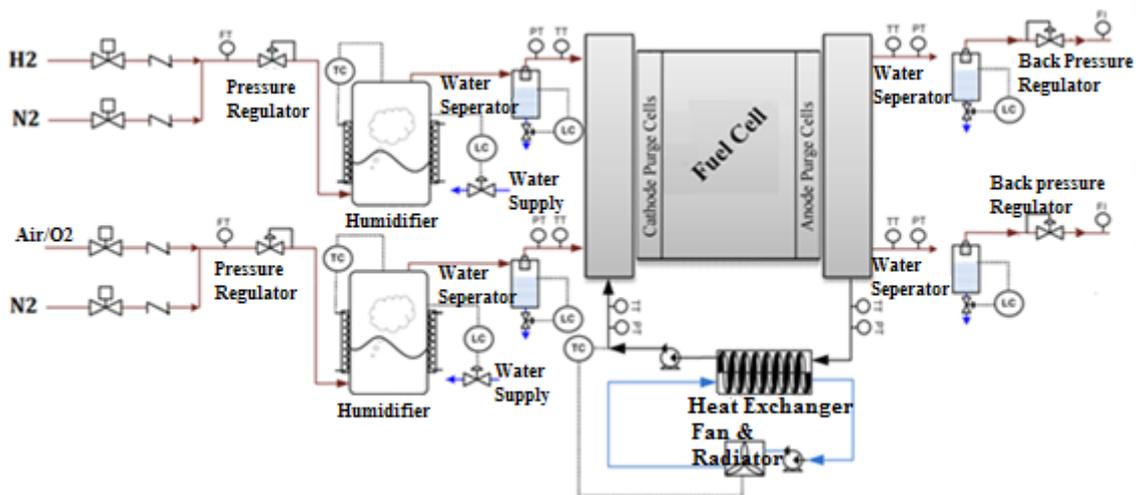


Fig. 4. Schematic representation of fuel cell test stand.

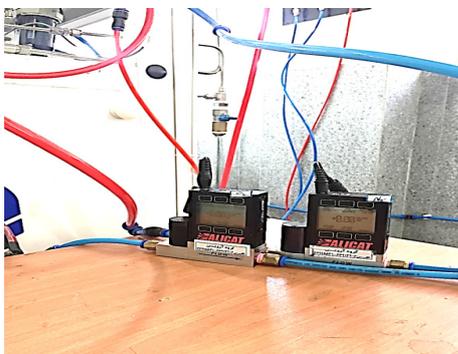


Fig. 5. Artificial air production system mass flow controllers.

In all of the tests, the dew point temperature and the pressure of the fuel and the oxidant were set to 65 °C and 1 barg, respectively. The cell temperature was set to 70 °C with the hydrogen and air stoichiometries of

1.5 and 3, respectively. Hydrogen and air stoichiometries were defined as follows:

$$St_{H_2} = \frac{Q_{H_2,inlet}}{Q_{H_2,consumption}} \tag{1}$$

$$St_{air} = \frac{Q_{air,inlet}}{C \times Q_{air,consumption}} \tag{2}$$

During the lifetime tests, all the mentioned parameters were held constant, while for the sensitivity tests different parameters related to the target test have been changed.

3. Results and Discussion

3.1. Operating Condition

After exploring the durability of the H₂/Air fuel cell, we determined that it generated lower power com-

pared to H₂/O₂ due to its greater sensitivity to the changes in operating conditions. The performance of this cell was evaluated by tuning different parameters as tabulated in Table 2. The range of variation and uncertainty of measured parameters are listed in Table 3.

Table 2. Test point matrix based on the fuel cell operating conditions (fuel cell Temperature = 70 °C).

Test#	Idens [A/cm ²]	Stoic.C [-]	DPT.Si.A [°C]	p.Si.A [barg]
			DPT.Si.C [°C]	p.Si.A [barg]
1	0.2	1.5 4 3 2	65	1
	0.4	1.5 4 3 2	65	1
	0.6	1.5 4 3 2	65	1
2	0.6	1.5	50 60 65 70 75 80	1
3	0.2	1.5	65	0.8 1 1.2
	0.4	1.5	65	0.8 1 1.2
	0.6	1.5	65	0.8 1 1.2

Table 3. Range of variation and uncertainty of measured parameters in tests.

Parameter	Range or Value	uncertainty
Current	0-300 A	±1 A
Voltage	0-4 V	±0.01 V
Molar Concentration of H ₂ tank	99.999 %	±0.005 %
Molar Concentration of O ₂ tank	99.995 %	±0.005 %
H ₂ pressure	0-300 kPa	±0.35 kPa
O ₂ Pressure	0-300 kPa	±0.35 kPa
H ₂ Volumetric flow rate	0-60 SLPM	±0.06 SLPM
O ₂ volumetric flow rate	0-30 SLPM	±0.03 SLPM
Cooling fluid Volumetric flow rate	0-35 LPM	±1 LPM
Inlet O ₂ stoichiometry	1-3	-
Inlet H ₂ stoichiometry	1-2	-
Relative humidity of inlet O ₂	0-100	±2 %
Relative humidity of inlet H ₂	0-100	±2 %
Fuel cell temperature	20-80 °C	±2 °C
Cooling fluid temperature	20-80 °C	
H ₂ gas temperature	15-80 °C	
O ₂ gas temperature	15-80 °C	

After a lifetime test, the effect of air stoichiometry on the performance of fuel was investigated. Fig. 6 shows the behavior of cell voltage in terms of stoichiometric changes of air from 1.5 to 4. At the start of the tests, the air stoichiometry was set to 4, and it was gradually reduced to 1.5. Fig. 6 shows that increasing the air stoichiometry gradually improved the fuel cell performance. Fig. 6 shows that low air

flow rates result in an oxygen starvation situation since the oxygen existing in the air flow has been consumed through the channel path. Also, a low air flow rate decreases the capability of water elimination, and thus, prevents oxygen from reaching the active sites of the MEA. Due to the reasons stated, the fuel cell performance decreases. However, low air flow rates increase the membrane's moisture re-

sulting in reduced electrical resistance and improved fuel cell performance. A high air flow rate increases the amount of water elimination and reduces the water content, which leads to increased electrical resistance. On the other hand, high air flow increases oxygen accessibility on the MEA, which improves the performance of the fuel cell. The air flow rate is said to be optimum when the positive effects of increased oxygen accessibility prevail over the negative effects of membrane drying. In all three current densities, the voltage changes are more predominant in higher current densities than the lower ones for air stoichiometries of less than 3 and a constant value in this range. In other words, the sensitivity of voltage to stoichiometric changes is more effective for high current densities. This stems from the fact that chemical reactions proceed at higher rates at high current densities, and higher amounts of water will be generated. At stoichiometries less than 3, the air flow rate is not enough to eliminate the generated water, and as a consequence, the voltage decreases with a higher slope.

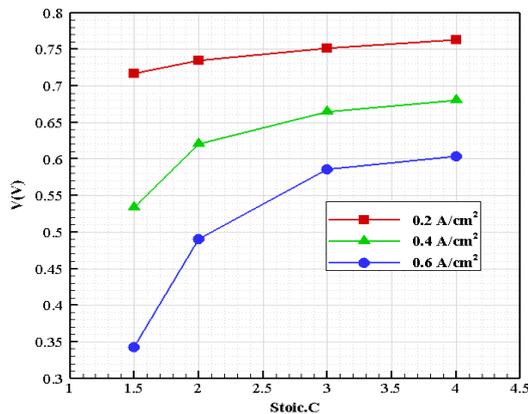


Fig. 6. Air stoichiometry effect on the fuel cell performance (fuel and air dew point temperature= 65 °C, fuel and air pressure= 1 barg, hydrogen St = 1.5).

The pressure is another parameter that has a significant effect on the performance of a fuel cell. Fuel cell parameters, such as the composition of the inlet gas and the permeability of the gas diffusion layer, may change with the reactant's gas pressure. In this test, the pressure varies from 1.2 to 0.8 barg. Since the saturation pressure is constant at a constant op-

erating temperature, as the pressure rises, the molar fraction of water vapor within the inlet gas decreases due to water condensation. Consequently, the percentage of oxygen molecules increases by increasing operating pressure. Fig. 7 illustrates the variation of voltage in terms of pressure. As the operating pressure rises from 0.8 to 1.2 barg, fuel cell performance also increases.

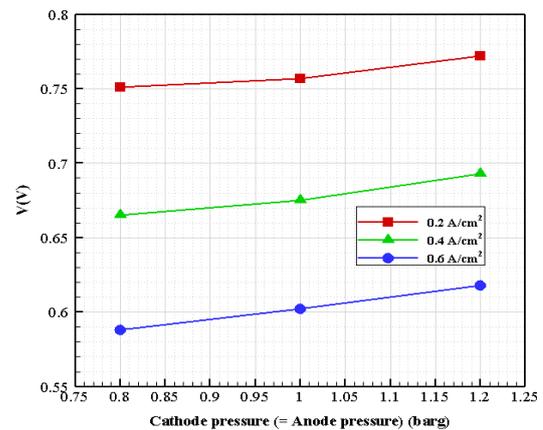


Fig. 7. Reactants pressure effect on the performance of a fuel cell (fuel and oxidant dew point temperature= 65 °C, air St=3, hydrogen St = 1.5).

The effect of the inlet gas temperature on the fuel cell performance is shown in Fig. 8. As can be seen, the fuel cell performance was improved by increasing the temperature to about 75 °C, which can be attributed to the enhanced penetration of the reactant gases, improved membrane conductivity, and higher reactivity of the reactant gases. Conversely, the vapor condenses easily at lower temperatures and blocks the channels of the flow field. This water condensation is also responsible for the flooding in the gas diffusion layer, which reduces the permeability of the reactant gas in the catalyst and gas diffusion layer and leads to serious drawbacks in the operation of the components and performance of the cell. In addition, higher temperatures, up to 75 °C, decrease the water content of the membrane, and adversely affect the proton conductivity. This consequently leads to a reduction in fuel cell performance at temperatures above 75 °C.

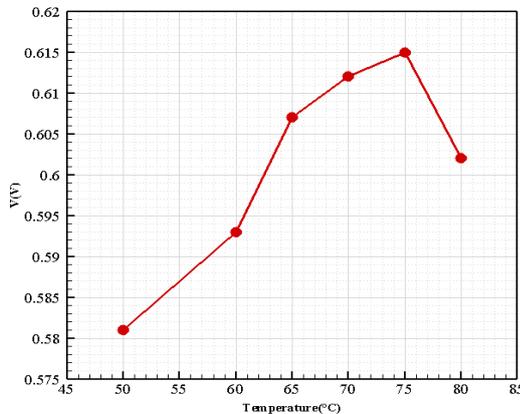


Fig. 8. Reactants dew point temperature effect on the performance of a PEMFC (fuel and air pressure= 1 barg, air St = 3, hydrogen St = 1.5).

3.2. Lifetime Fuel Cell Testing

The lifetime test of the fuel cell was started with the fuel and oxidant stoichiometries of 1.2 and 2.5, respectively. This stoichiometric condition resulted in a low and highly fluctuating cell voltage, probably due to the accumulation of water within the flow field and blockage of the channels. To overcome this problem, the stoichiometry of the hydrogen was initially increased to 2 and then decreased to 1.5. Moreover, the stoichiometry of air was set to the constant value of 3. As can be clearly seen in Fig. 9, as the changes were applied to the operating condition, the initial low voltage partially increased, but the trend of fluctuations remained unaltered. To solve this problem, the temperature of the inlet gas was investigated. Fig. 10 shows the voltage decay of the cell as well as the effect of the inlet gas temperature on the voltage oscillations. During the tests, the fuel cell was operated for approximately 9 hours and was then turned off for about 14 hours, which caused the cell to recover its initial voltage,

Fig. 10. A high voltage oscillation was observed when the gas initially entered the fuel cell at 80 °C. Some of the moisture content of the inlet gas was condensed after entering the fuel cell due to the lower temperature of the cell. This condensation blocked the channels and led to a decrease in the active surface area and the obtained voltage. The continuous process of generation and elimination of water within the channels of the flow field eventually caused the voltage fluctuation. The inlet gas temperature was reduced to 70 °C as a workaround for these fluctuations, and the voltage fluctuation decreased. However, after decreasing the temperature to this value, the overall voltage of the cell increased to some extent, which can be attributed to the increase in catalytic active surface area over time. The temperature of the inlet gas was then reduced to 65 °C, which decreased the voltage fluctuation due to decreased water formation in the channel. To ensure the correct operation, the temperature was increased until the oscillation reappeared at 70 °C. Finally, the test continued at 65 °C, and as shown in Fig. 10, the oscillations were greatly reduced.

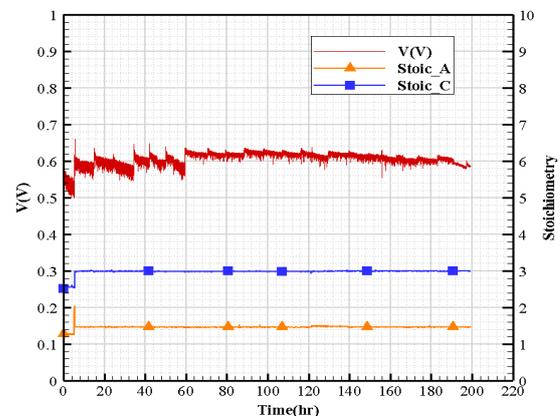


Fig. 9. Effect of reactants flow rate on the fluctuation of voltage in a durability test with constant load.

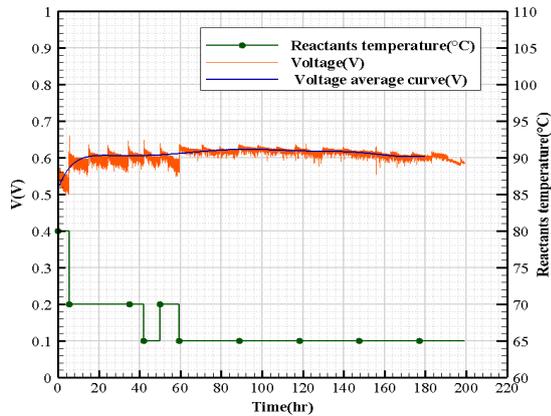


Fig. 10. Impact of reactants dew point temperature on fluctuation of voltage in a durability test with constant load.

To investigate the possibility of employing this fuel cell for other applications, a test was performed for 80 hours with the same final operating conditions employed in the H₂/Air durability test, but this time the air was replaced by oxygen on the cathode side. The nitrogen gas flow rate was set to zero through the MFC, and the test was run for approximately 80 hours. Finally, the results were compared with the H₂/Air test. As shown in Fig. 11, due to the lower flow rate of oxygen in comparison to the flow rate of air (approximately 20%), less moisture and water entered the cell, which results in less voltage and power fluctuation than the H₂/Air cell. The main reason for the lower generated power of the H₂/Air cell compared to the H₂/O₂ counterpart stems from the

concentration of O₂ gas within the oxidant. In the H₂/Air cell the oxygen was diluted with nitrogen gas and did not have enough accessibility to the active sites of the catalyst due to the higher flow rate of oxidant. Also, the presence of nitrogen remaining from the previous purge cycle within the GDL pores causes the voltage to rise at the beginning of the H₂/O₂ test, which gradually releases and enhances the voltage.

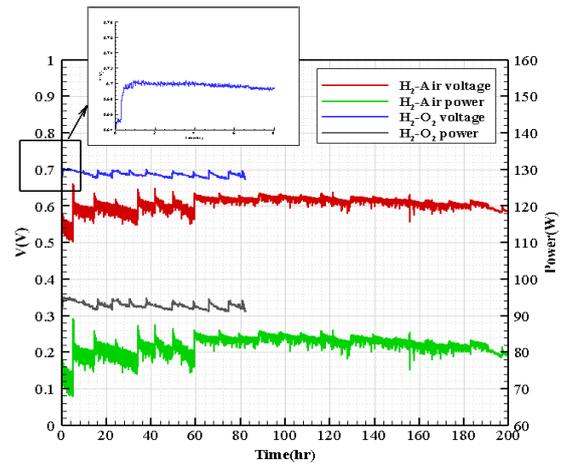
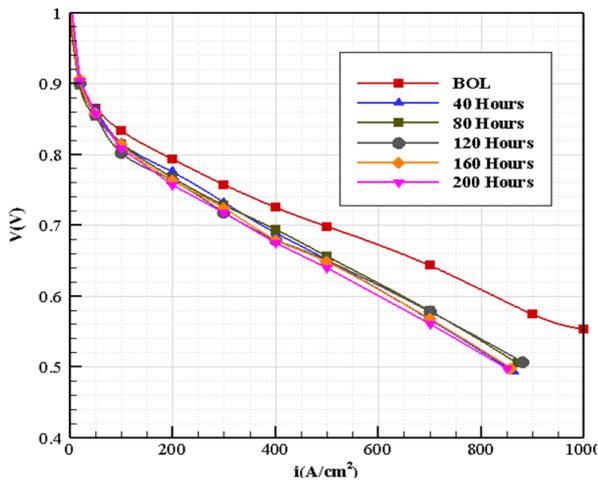
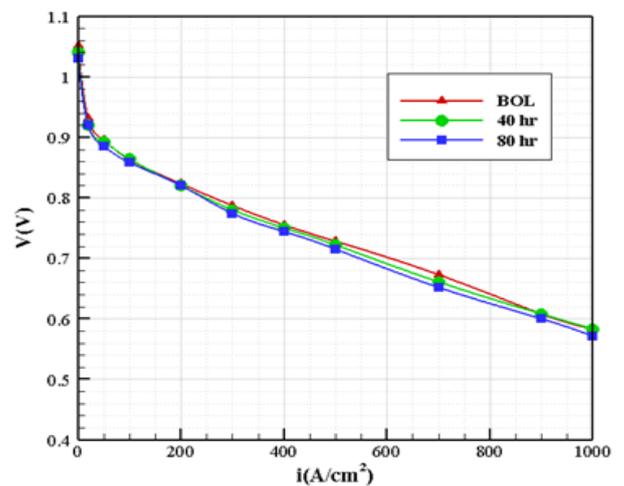


Fig. 11. Comparing two oxidants of a PEMFC.

The polarization curve was obtained at the beginning of the lifetime test and continued in 40 hours intervals. As shown in Fig. 12, the performance loss of the fuel cell over time is evident for both air and oxygen oxidants, but the loss rate in the air are bigger than the O₂ oxidant. It should be noted that during the lifetime test, the polarity diagrams of the fuel cell should be similar in order to make a fair comparison.



(a)



(b)

Fig. 12. Polarization curve for two oxidants during the lifetime test, a) Air and b) O₂.

4. Conclusions

In this work, a multi-functional fuel cell has been assembled to be capable of using both air and oxygen with respect to the target application. Lifetime testing was carried out for the H₂/Air fuel cell, whose power generation capability is highly sensitive to operating conditions in comparison to its H₂/O₂ counterpart. The cathode stoichiometry was set to 3 as the optimal value for the H₂/Air fuel cell with an inlet gas pressure of 1.2 barg and temperature of 75°C. While exhibiting the highest performance at this temperature, the inlet gas temperature was finally set to 65 °C because of severe voltage fluctuations. The tests were performed on the fuel cell using two different oxidants (air and pure oxygen), and the same operating conditions were applied during the 80 hours of tests. In this study, the effects of pressure, varied from 1.2 to 0.8 barg, were investigated. As the pressure rose, the molar fraction of water vapor within the inlet gas decreases due to water condensation. Consequently, the fuel cell's performance increased. The results confirmed the possibility of employing this fuel cell with two different oxidants for different applications. Although it produced 15% less power than the pure oxygen oxidants and showed more fluctuating behavior due to higher gas flow rate and humidity, the results of the fuel cell with air oxidant were satisfactory.

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