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Parameters Affecting the Thermodynamic Efficiency of PEM Single Cell and Stack of Cells (Two Cells)

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Abstract

In this work, thermodynamic efficiency of individual cell and stack of cells (two cells) has been computed by studying the variation of voltage produced during an operation time of 30 min as a result of the affected parameters:- stoichiometric feed ratio, flow field design on single cell and feed distribution on stack of cells. The experiments were carried out by using two cells, one with serpentine flow field and the other with spiral flow field. These cells were fed with hydrogen and oxygen at low volumetric flow rates from 1 to 2 ml/sec and stoichiometric ratios of fuel (H_2) to oxidant (O_2) as 1:2, 1:1 and 2:1 respectively. The results showed that the highest voltage and efficiency can be obtained for the stoichiometric ratio of 1:2, while the ratio of 2:1 produced the lowest voltage and efficiency. Also the best results were obtained with the serpentine flow pattern after comparing with the spiral flow pattern in a single cell. Likewise it was proved that the voltage and efficiency are maximized when using the stoichiometry of 1:2, besides that the parallel feed connection of the stack of cells produced much power than the series connection.

Keywords: Thermodynamic efficiency, Proton Exchange Membrane Fuel Cell (PEMFC), Stoichiometric ratio, flow field design, feed distribution.

Introduction

At the beginning of the 20th century, the equipment that converted chemical energy into electrical energy became more urgent due to the increase in the demand of electricity to minimize the energetically reliance on fossil fuels and lessen dangerous emissions into the atmosphere. Fuel cell (FC) has high electrical efficiency compared to other sources [1]. Nowadays fuel cells are very useful power sources than before, and they have the ability to satisfy the global power demands [2]. Fuel cells are electrochemical devices that utilize hydrogen (H_2) together with oxygen or (oxygen from air), to generate electricity and water. However there are many variants of this process, basing on the type of fuel cell and the fuel used [3]. Different types of fuel cells which could be recognized by power produced into a range between few watts and megawatts. They are Proton Exchange Membrane Fuel Cell (PEMFC), Direct Methanol Fuel Cell (DMFC), Alkaline Fuel Cell (AFC), Phosphoric Acid Fuel Cell (PAFC), Molten Carbonate Fuel Cell (MCFC), Solid Oxide Fuel Cells (SOFC). All types of fuel cell are typically divided due to the nature of the electrolyte used [4].

Proton exchange membrane (PEM) fuel cell has many advantages such as low operating temperature, high power density, rapid startup, as well as excellent reliability and durability over other types of fuel cells [5]; it combines as very active unit the electrodes and the electrolyte. This design is well known as Membrane Electrode Assembly (MEA) [6]. During this process, the protons diffuse through the electrolyte membrane to the cathode, where they recombine to form hydrogen gas and react with oxygen to produce water using the simple redox reaction mechanism

Anode: $H_2 \rightarrow 2H^+ + 2e^-$ …(1)

Cathode: $1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O$ …(2)

Net Reaction: $H_2 + 1/2O_2 \rightarrow H_2O + heat$ …(3)

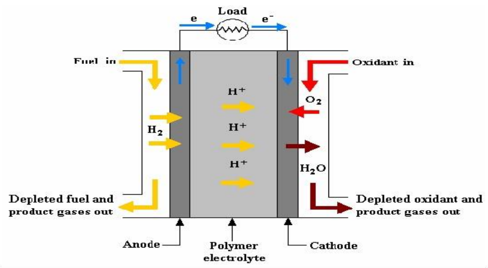

Fig. 1: PEM fuel cell design

Like any power generated device, PEMFC performance usually is affected by many parameters like pressure, temperature, relative humidity and feed stoichiometric ratio [7].

The ratio of amount of actual flow rate of reacted components at inlet of PEMFC to the amount is being consumed by the redox reaction is called stoichiometry. The product is water and in order to enhance the reaction rate, this water must be carried out from the cell by increasing the flow rate particularly on the cathode side where the chemical reaction took place and the water is continuously formed [8]. On the other hand, the purity of both supplied feed is extremely significant as its effectiveness on the operative behavior of cell. If hydrogen and oxygen are pure, then the needed flow rates are the same as the consumption and they are usually at low pressure. But when the feed of gases are not pure (oxygen from air) or impurities are existed with hydrogen in the feed, then nitrogen could be used to purge these accumulative materials or by the increasing of gas pressure to exclude these contaminants. Because of the required flow feed is considered a design variable, this flow rate is related to the possible efficiency of reaction .the extremely higher flow rate the lower efficiency because of wasted hydrogen and when the flow rate is too low, the performance may suffer[9].

Another design parameter is the flow field pattern. Its impact on cell performance can be summarized by several functions like providing flow channels for reactive gases to their respective anodic and cathodic electrode surfaces, providing path for the removal of water generating from electrochemical reactions, considering as a mechanical support for the electrodes, serving as a current collector, electronically connecting one cell to another in a stack and acting as a physical barrier to prevent reactants and coolant fluids from mixing [10].

For conventional types of fuel cells like SOFC, DMFC and PEMFC, an individual cell produces 1 volt under operational conditions where the design of a single cell is usually used for lab testing purposes. Therefore to increase power for practical application, it is more convenience for utilizing multiple fuel cells stacked together than to use a single cell [11]. Because of kinetic losses a single fuel cell has low power density. Consequently, it was important to reduce these losses so as the design purpose of stack will be beneficial and more efficient [12]. For a fuel cell stack, beside these losses in every cell, the non-uniform distribution of fuel and oxidant to the multiple fuel cells can also cause decrease of the maximum available power output from the stack [13].

Marston has studied the design, fabricating and assembling of a PEMFC and evaluating cell efficiency by the variation of the H2/air feed ratio and acquired a representative plot of the fuel cell voltage as a function of time [14]. Cai, Hu, and Zhang have found that the cell performance and MEA resistance varied a little when utilizing thin membrane while the anode humidity changed from saturated to dry. The effectiveness of the cathode humidity was serious on the cell behavior [15]. Guvelioglu and Stenger have presented a mathematical model by studying cell efficiency for different hydrogen flow rates, air flow rates and humidification levels. They have revealed the sensitivity of current density to both gases flow rates as well as their relative humidity [16]. Jang, Chiu, Yan and Sun studied the behavior of single cell and stack of cells due to four effective key parameters (gas humidification temperature, cell temperature, assembled torsion, and gas flow rate). They revealed that the center cells have lower efficiency than the cells at both sides of the stack. Also they found that the efficiency did not change with maximizing the anode gas stoichiometric ratio but they saw the increase action with the increase in the cathode gas stoichiometric ratio [17]. Hsieh, Huang and Her found that the interdigitated flow design was having the largest pressure drop as much as the water accumulation at the early phase of ≤ 30 min in comparison to the rest of flow design shapes [18]. Higier and Liu, tried to optimize the serpentine flow design by developing a new technique in a house and separately measure the current density under the land and channel on different serpentine flow designs. Every single flow field was tested with variable conditions and that showed the flow field with thinner land and channel was better [19]. Taccani and Zuliani focused on studying the influence of flow field geometry of high temperature (120-180 ˚C) Polybenzimidazole (PBI) PEM composite bipolar plates on the fuel cell behavior. With three different channel geometry (two serpentine and one parallel), it was shown that the serpentine flow pattern is better [20]. Berning and Kær studied the possibility of operating PEMFC at low stoichiometric flow rate ratios using interdigitated flow pattern with dry feeds. It was shown when utilizing the stoichiometry (ξ c =1.2-1.5) at the cathode and as low as possible at the anode (ξ a =1) at ambient pressure leads to cell drying, and to avoid this case was to operate at or below 70 C° [21]. Yadava, Sahu and other researchers studied PEMFC performance with different process parameters like temperature, pressure, relative humidity and feed flow rates. It was observed that increasing in the above parameters will enhance the fuel cell performance [22]. Liu, Li and Wang investigated the effects of different gas flow field design on PEMFC behavior. It was proven that serpentine flow channel is the best among all other flow designs [23].

In this work, thermodynamic efficiency of individual fuel cell and stack of fuel cells (two cells) has been computed by studying the variation of voltage produced during an operation time of 30 min as a result of the effected parameters like stoichiometric feed ratio, flow field design on single cell and feed distribution on stack of cells.

Experimental Work

1. Operating Variables

In this study the following variables were taken into consideration:

1. Number of PEM fuel cells (one or two fuel cells).
2. Volumetric flow rates of the fuels (Hydrogen and Oxygen) fed to the fuel cell.
3. Fuel ratios (Hydrogen / Oxygen).
4. Type of flow field design (spiral or serpentine).
5. Type of electrical fuel cell connections (series and parallel).

2. Experimental Rig

The experimental rig is shown in Figure 2 and it consisted of the following:

1. A PEM fuel cell assembly which consisted of the following components:

a) Membrane Electrode Assembly (MEA) which consisted of a polymer membrane type Nafion 117 with active area of D = 8.8 cm and a total area of D = 10 cm.

b) Two catalytic electrodes (Anode and cathode) which consisted of carbon loaded with 0.3 mg/cm² PtC (40%) for each electrode.

c) Two gas diffusion layers.

d) Two carbon end plates (10cm in diameter) with serpentine and spiral gas flow fields with groove width and depth of 1mm as shown in Figures 3 and 4 respectively.

e) Two current collectors made from copper of 99.5% purity cut in circular shape.

f) Two Teflon end plates with a canal for the rubber gasket. The Teflon end plates sandwiches the above components with stainless steel bolts and nuts.

2. Two cylinders one to store the hydrogen fuel and the other for the oxygen fuel.
3. Two cylinder regulators one for the hydrogen cylinder and the other for the oxygen cylinder.
4. Six calibrated flow meters. Each three flow meters are connected in parallel to achieve the desired gas (hydrogen or oxygen) flow range.
5. PVC tubes to transport the fuel gas (hydrogen or oxygen) from the cylinder to the three parallel flow meters then to the PEM fuel cell.
6. Two inside feeds for hosing, PVC tubes inside the Teflon end.
7. Voltmeter with wire leads to read the voltage output of the PEM fuel cell.

Fig. 2: Experimental rig for PEM fuel cell thermodynamic study

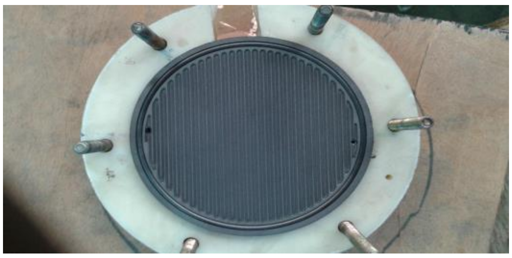

Fig. 3: Serpentine Carbon end with a rubber gasket fitted on top of a copper collector inside a Teflon end

Fig. 4: Spiral Carbon end with a rubber gasket fitted on top of a copper collector inside a Teflon end

3. Operation of PEM Fuel Cell

Once the fuel cell was found to be functional, several tests were performed to determine its capabilities and enhance understanding of its operation. For all experiments, the behavior of produced cell voltage versus time was studied by using the following operating conditions:

1. Atmospheric pressure (1 atm).
2. Low operating temperature (ambient temperature).

3.1. Single Fuel Cell

A single fuel cell was assembled with membrane electrode assembly and carbon bipolar plates with spiral flow pattern. This fuel cell was operated with three different stoichiometric feed flow rate ratios. The stoichiometry ratios (SR) were 1:2, 1:1 or 2:1 for hydrogen to oxygen. The gas flow rate was from 1 to 2 ml/sec. The voltage produced was recorded for each one minute operation during a time of 30 minutes.

Another single fuel cell was assembled with the same assembling compositions but this time with serpentine flow pattern. This fuel cell operated with same three different stoichiometric feed flow rate ratios. The stoichiometry ratios (SR) were 1:2, 1:1 or 2:1 for hydrogen to oxygen. The gas flow rate was from 1 to 2 ml/sec. The voltage produced was recorded for each one minute operation during a time of 30 minutes. For each one of the two above fuel cells the power voltage begin rising till it reached the maximum value, after that the produced voltage begin to fall down until it reached the stable value through time evaluated by a stop watch through the experiment.

3.2. Fuel Cell Stack (Two Cells)

Two fuel cells were stacked together each fuel cell was assembled with membrane electrode assembly and carbon plates with serpentine and spiral flow patterns. These fuel cells in the stack were operated with three different stoichiometric feed flow rate ratios. The stoichiometry ratios (SR) were 1:2, 1:1 or 2:1 for hydrogen to oxygen. The gas feed flow rate was from 1 to 2 ml/sec. The voltage produced was recorded for each one minute operation during a time of 30 minutes. The two fuel cells in the stack were connected in series. Another type of operation for the stack of fuel cells was achieved by connecting these cells in parallel. For stack of fuel cells the power voltage begin rising till it reached the maximum value, after that the produced voltage begin to fall down until it reached the stable value through time evaluated by a stop watch through the experiment.

Results and Discussion

1. Voltage

For all experiments and as illustrated in Figures 5 to 8, the voltage produced due to cell or cells operation was suddenly rising and reaching to maximum value through 1 min and then falling down till it reached the steady state in the end of 30 min. This is by virtue of the kinetic potential losses, especially the sluggish oxygen reduction reaction which causes the potential loss or degradation in voltage produced at standard conditions even when pure oxygen gas was used. On the other hand, the hydrogen oxidation contributes very little to the polarization under normal operating conditions and has therefore gained less attention than the slowest process of oxygen reduction [24].

2. Effect of Stoichiometric Ratio of Feed Flow Rates on Fuel Cell Performance

The effects of gas stoichiometric ratio on the performance of individual cell and cell stack are presented in Figures 5 to 8. Data values of voltage that were obtained as a result of feed stoichiometric ratio $H_2/O_2=0.5$ were the highest values during the time of operation compared to other feed ratios while for the ratio $H_2/O_2=2.0$, the results were the lowest values. This was due to water produced from chemical reaction, specifically on cathode side which affected the reaction rate by virtue of being a barrier to the oxygen gas molecules to reach the active sites on catalytic layer (CL), the location where the chemical reaction would take place. The decreasing in produced voltage when utilizing feed ratio ($H_2/O_2=2$), was a result of limitation in O_2 mass transport through the gas diffusion layer (GDL) and the transportation of excess water to the flow channels. Liquid water in the GDL was frequently assumed to be the major cause of mass transport limitations [25].

3. Effect of Flow Pattern of Feed on Fuel Cell Performance

Two flow patterns spiral and serpentine were used when operating the PEM fuel cell. As can be noticed from Figures 5 and 6 the serpentine flow pattern gave the highest produced voltage of 977mVolt for single fuel cell with H2/O2 fuel ratio of 0.5 while the spiral flow pattern gave a value of 885mVolt for the same conditions. It was obviously noticed that the serpentine channel design was favorable because of its capability to balance water removal and to control pressure drop to avoid water flooding and membrane dehydration as shown by [26, 27]. For other fuel cell with spiral flow pattern, it was found that there were tiny drops of water remaining in the flow channels when the cell was opened. It indicated that inefficient water removal was the justification of low power output.

4. Effect of Feed Distribution on Stack (Two Cells) of Fuel Cell Performance

The performance and power produced from two stacked cells by means of connection type was studied. As can be noticed from Figures 7 and 8, the highest produced voltage was 1860 mvolt for stack with parallel feed and H_2/O_2 fuel ratio of 0.5. It was clearly shown that the stack of cells with the distributed feed in a parallel connection was preferred than series because each individual cell was being supplied with a uniform amount of fuel and oxidant compared with series connection. Otherwise a non-uniform feed supply will lead to increasing non uniform chemical reaction and that will minimize the power of the entire stack [28, 29].

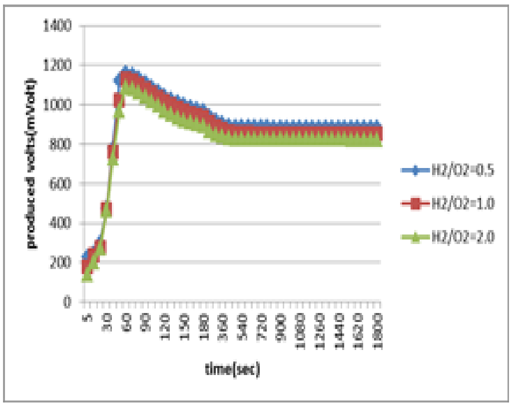

Fig. 5: Voltage produced with time from the operation of single PEM fuel cell with three stoichiometric ratios (1:2, 1:1and 2:1) and spiral flow field pattern

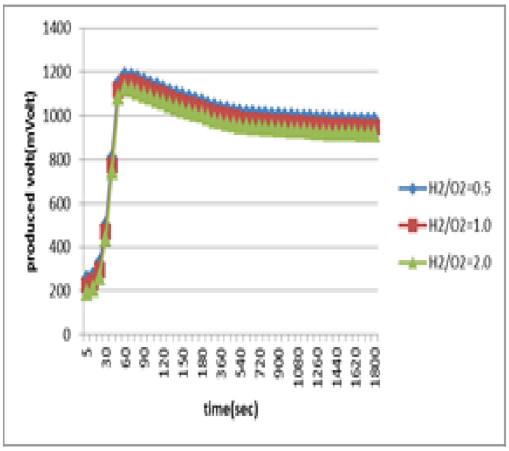

Fig. 6: Voltage produced with time from the operation of single PEM fuel cell with three stoichiometric ratios (1:2, 1:1and 2:1) and serpentine flow field pattern

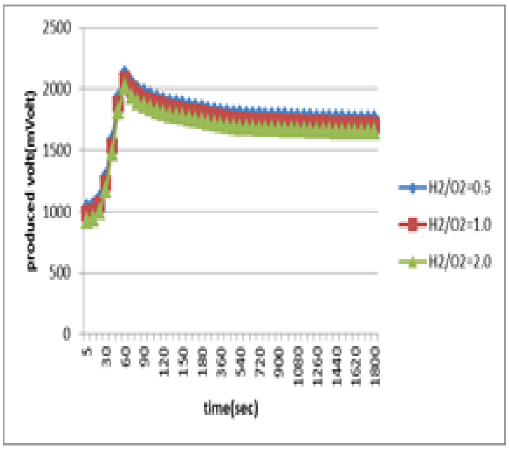

Fig. 7: Voltage produced with time from the operation of PEM fuel cell stack connected in series with all three stoichiometry

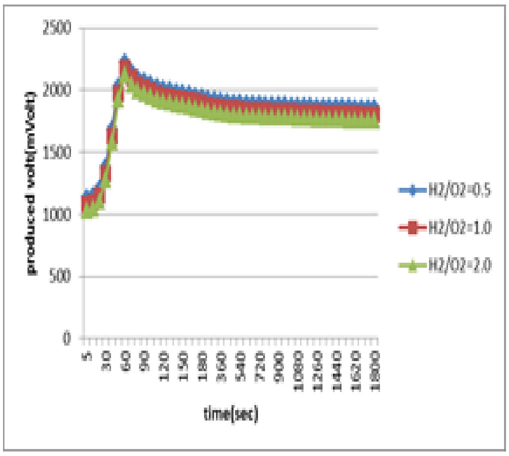

Fig. 8: Voltage produced with time from the operation of PEM fuel cell stack connected in parallel with all three stoichiometry

5. Thermodynamic Efficiency of PEM Fuel Cell

The efficiency of a PEM fuel cell was determined .Both the enthalpy of chemical reaction and Gibbs free energy are at standard state. For PEM fuel cell the overall reaction was:

H_2 (g) + 1/2O_2 (g) →H_2O (l), with ΔG° = -237.18 kJ/mole and ΔH° = -285.64 kJ/mol [30]. Theoretical efficiency, η of PEM fuel cell based thermodynamic quantities was:

$$\eta = \Delta G^\circ / \Delta H^\circ = -237.18/-285.64 = 83\% \quad \ldots(4)$$

The second law of thermodynamics shows this is the maximum efficiency that can be obtained. The anode and the cathode half-reactions in the PEMFC are:

$$H_2 \rightarrow 2H^+ + 2e^- \quad E^\circ = 0.000\ V\ (anode)$$
$$1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O \quad E^\circ = 1.229\ V\ (cathode) \quad \ldots(5)$$

Thus the overall reaction potential equals 1.229 V, which is the maximum voltage possible [30]. The observed efficiency of the fuel cell which was defined as:

$$\%\eta = 100*E/E^\circ \quad \ldots(6)$$

Where E is the measured cell potential (produced voltage) and E° is the theoretical voltage, or 1229 mVolt for the PEM cell. Table 1 lists the maximum and average efficiency of all fuel cell operations performed in this study using Equation 6.

As can be noticed from Table 1 the highest average efficiency is for the operation of single fuel cell with serpentine flow pattern and stack of cell with parallel connection with the H_2/O_2 fuel ratio of 0.5.

Table 1: The maximum and average efficiency of all fuel cell operations

Ratio	Flow Pattern	No. of Fuel cells	Type of connection	Max %η	Ave. %η
0.5	spiral	1	Non	94	72
1.0	spiral	1	Non	92	70
2.0	spiral	1	Non	89	67
0.5	serpentine	1	Non	96	79
1.0	serpentine	1	Non	94	77
2.0	serpentine	1	Non	91	75
0.5	Serpentine and spiral	2	Series	86	72
1.0	Serpentine and spiral	2	Series	84	70
2.0	Serpentine and spiral	2	Series	82	67
0.5	Serpentine and spiral	2	Parallel	91	76
1.0	Serpentine and spiral	2	Parallel	88	74
2.0	Serpentine and spiral	2	Parallel	86	72

Conclusions

1. Degradation in voltage is a result of kinetic over potential.
2. For low feed flow rates, the increase in the amount of oxidant relative to the fuel stoichiometry will enhance the performance of PEM fuel cell at low pressure and temperature.
3. The flow field design was an effective parameter in fuel cell operation, where serpentine flow pattern was more favorable than spiral one.
4. Proper feed distribution in stack of cell will enhance the requirement of high efficiency.

Abbreviation

FC=Fuel cell
PEMFC= Polymer Electrolyte Membrane Fuel Cell.
DMFC= Direct Methanol Fuel Cell.
AFC=Alkaline Fuel Cell
PAFC= Phosphoric acid fuel cell
MCFC=Molten carbonated fuel cell
SOFC =Solid oxide fuel cell MEA= Membrane Electrode Assembly
SR=Stoichiometric Ratio
CL=Catalytic Layer
GDL=Gas Diffusion Layer

Nomenclature

η =Thermodynamic efficiency
ξ=Feed stoichiometric ratio
ΔH°=Change in enthalpy at std. conditions
ΔG°=Change in Gibbs free energy at std. conditions

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