Thermodynamic Study of Operation Properties Effect on Polymer Electrolyte Membrane Fuel Cells (PEM)

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Abstract: The thermodynamic analysis of PEM fuel cell energy production depends on the entropy and enthalpy of reaction with the changing of the operating temperatures that ranges between 50 and 100°C, the electrical work done will be equal to the Gibbs free energy released. This paper presents a mathematical model of PEM fuel cells, based on physical-chemical procedures of the phenomena occurring inside the fuel cell, and it was theoretically studied the performance at different operation variables and conditions. The C++ program is designed to calculate all thermo-chemical parameters, i.e. enthalpy of formation, Gibbs free energy, work and efficiency for any type of fuel cells. The results are plotted as a function of fuel cell operating temperature. The results shows that the highest value of Gibbs energy is at the lowest operating temperature, and decreases gradually with increasing the temperature, the output voltage is determined by cell’s reversible voltage that arises from potential difference produced by chemical reaction and several voltage losses that occur inside a cell. In addition the results showed that the efficiency of this type of the fuel cells is much higher than the ideal Carnot’s efficiency, it changes between 82% to 85% depends on temperature operation. The heat output (required heat) from the fuel cell increases with increasing the operating temperature, this heat is used for many thermal applications such as buildings space heating.

الملخص: يعتمد التحليل الدينيميكى الحراري لإنتاج طاقة خلايا الوقود ذات غشاء الفصل البوليمرى على الإنتروبيا والمحتوى الحراري للتفاعل، مع تغير PEM مع تغير الخصائص التشغيلية لخلايا الوقود.

دراسة الديناميكا الحرارية لتأثير الخصائص التشغيلية لخلايا الوقود ذات غشاء الفصل البوليمرى

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Solar Energy and Sustainable Development, Vol. 7, No. 1, Jun. 2018
1. INTRODUCTION

The fuel cells are primarily designated for usage in a specific type of application such as vehicular transportation, stationary power energy generation, space application and mobile electronic power source. Fuel cells technology still young technology in Libya, and there is no any applications in this country, although, it can be used in a wide range of applications, including transportation, material handling, stationary, portable, and emergency backup power applications. Fuel cells have several benefits over conventional combustion-based technologies currently used in many power plants and passenger vehicles. Fuel cells can operate at higher efficiencies than combustion engines, and can convert the chemical energy in the fuel to electrical energy with efficiencies of up to 60%. Fuel cells have lower emissions than combustion engines. Hydrogen fuel cells emit only water, so there are no carbon dioxide emissions and no air pollutants that create smog and cause health problems at the point of operation. Also, fuel cells are quiet during operation as they have fewer moving parts.

Polymer electrolyte membrane (PEM) fuel cells (also called proton exchange membrane fuel cells PEMFC) are believed to be the best type of fuel cell as the vehicular power source to eventually replace the gasoline and diesel engines. PEM fuel cells use a solid polymer membrane (a thin plastic film) as the electrolyte. This polymer is permeable to protons when it is saturated with water, but it does not conduct electrons [1,2].

The fuel for the PEMFC is hydrogen and the charge carrier is the hydrogen ion (proton). At the anode, the hydrogen molecule is split into hydrogen ions (protons) and electrons as shown in Figure 1. The hydrogen ions permeate across the electrolyte to the cathode while the electrons flow through an external circuit and produce electric power. Oxygen, usually in the form of air, is supplied to the cathode and combines with the electrons and the hydrogen ions to produce water, [1, 3-6].

For a PEMFC, the electrochemical reactions are:

**Cathode:** \( \frac{1}{2}O_2(g) + 2H^+_i(aq) + 2e^- \rightarrow H_2O(l) \)

**Anode:** \( H_2(g) \rightarrow 2H^+_i(aq) + 2e^- \)

**Overall:** \( H_2(g) + \frac{1}{2}O_2(g) \rightarrow H_2O(l) \)

Figure (1). Polymer Electrolyte Membrane Fuel Cell.

**Keywords:** Operating temperature; Enthalpy of reaction; Gibbs free energy
The operating temperature of PEMFC is less than 100°C, which allows rapid start-up. These traits and the ability to rapidly change power output are some of the characteristics that make the PEMFC the top candidate for automotive power applications. Low temperature operation allows them to start quickly (less warm-up time) and results in less wear on system components, resulting in better durability. However, it requires that a noble-metal catalyst (typically platinum) be used to separate the hydrogen’s electrons and protons, adding to system cost. The platinum catalyst is also extremely sensitive to CO poisoning, making it necessary to employ an additional reactor to reduce CO in the fuel gas if the hydrogen is derived from an alcohol or hydrocarbon fuel, [2, 3, 7].

Other advantages result from the electrolyte being a solid material, compared to a liquid. The sealing of the anode and cathode gases is simpler with a solid electrolyte, and therefore, less expensive to manufacture. The solid electrolyte is also more immune to difficulties with orientation and has less problems with corrosion, compared to many of the other electrolytes, thus leading to a longer cell and stack life. PEM fuel cells are used primarily for transportation applications and some stationary applications.

One of the disadvantages of the PEMFC for some applications is that the operating temperature is low. Temperatures near 100°C are not high enough to perform useful cogeneration, [3,4].

The main objectives of this work are to perform a thermodynamics and thermo-chemical analysis of fuel cells to study and described the basic characteristics of the PEM fuel cell system and analyze their performance. The use of the first and the second laws of thermodynamics allow a simple description of a reversible fuel cell. Where the mathematical model for this type of fuel cell will include the follows:

1. Determine of the main chemical reaction equations of the fuel cell.
2. Selection of operating conditions of fuel cell
3. Calculation of the standard fuel cell electromotive force.
4. Calculation of the maximum work output, heat output and thermal efficiency.

The proposed model is based on electrochemical engineering fundamentals and the following assumptions will be used:

- All calculations in this paper are reversible, steady state, steady flow, and pure gases
- The C++ is modeled to be applied on any type of fuel cells.
- The fuel is H₂ gas or hydrocarbons or CO and the oxidant is O₂.
- The conversion of energy occurs isothermally and in constant volume.
- Temperature and pressure are uniform along the electrodes.

2. THERMODYNAMICS AND ELECTROCHEMICAL ANALYSIS OF FUEL CELLS

The thermo-chemical and electrochemical analysis of PEM fuel cell can be calculated by chemical parameters balance and energy balance of the fuel cell, with analyze their chemical reaction.

2.1. Energy Balance of Fuel Cell

The energy balance around the fuel cell as shown in Figure 2, is based on the energy absorbing/releasing processes (e.g., power produced, reactions, and heat loss) that occur in the cell. As a result, the energy balance varies for the different types of cells because of the differences in reactions that occur according to cell type, [5,6].

![Figure (2). Fuel cell energy balance](image-url)
In general, the cell energy balance states that the enthalpy flow of the reactants entering the cell will equal the enthalpy flow of the products leaving the cell plus the sum of three terms: (a) the net heat generated by physical and chemical processes within the cell, (b) the dc power output from the cell, and (c) the heat loss from the cell to its surroundings, [6].

2.2. Chemical Balance

The chemical balances for the reactions occurring inside the fuel cell are identical:

$$\left\{ \text{H}_2 + \frac{1}{2}\text{O}_2 \right\}_{\text{Reactants(r)}} \rightarrow \left\{ \text{H}_2\text{O} \right\}_{\text{Products(p)}}$$  

The chemical processes within the CV are governed by the First law of thermodynamics, i.e.

$$\frac{dE_{CV}}{dt} = \dot{Q}_1 - \dot{W} + \sum_p (n_{\text{f}}\bar{e}_p) - \sum_r (n_{\text{f}}\bar{e}_r)$$  

The enthalpy includes the enthalpy of formation, thermal enthalpy, as well as the kinetic and potential energies wherever appropriate, [6,7,8].

Assumptions:

1. NKEPE (Neglect effects of Kinetic and Potential Energies), then $\bar{e}_r = \bar{h}$
2. For SSSF processes: $\frac{dE_{CV}}{dt} = 0$ [24].

After applying assumptions, the relation obtained is:

$$\dot{Q}_1 - \dot{W} = \left[\left( n_{\text{f}}\bar{h}_{1,0}\right)_b + \left( n_{\text{f}}\bar{h}_1 + n_{\text{f}}\bar{h}_b \right)_b \right]$$  

Where, $n_{\text{f}}\text{H}_2 = 1, n_{\text{f}}\text{O}_2 = 1$, and $\bar{h}_{1,0}, \bar{h}_1, \bar{h}_b$ from above chemical reaction (1). Dividing the last equation by $n_{\text{f}}\text{H}_2$ and simplifying the resultant expression, the relation obtained is:

$$\dot{q}_1 - \dot{w} = \left( \bar{h}_{1,0}\right)_b + \left( \bar{h}_1 + \bar{h}_b \right)_b = \bar{h}_{RF}$$  

The reaction in the fuel cell produces both heat transfer and work. Solving (4) for the electrical work produced by the fuel cell per unit molar flow rate of fuel yields:

$$\bar{W}_{\text{FC, elec}} = \dot{q}_1_{\text{FC}} \cdot \bar{h}_{RF}$$  

2.3. Entropy Balance

Entropy balance for chemical reaction for a generic rate of heat transfer $\dot{Q}_1$ crossing the system boundary at $T_1$ is:

$$\frac{dS}{dt} = \frac{\dot{Q}_1}{T_1} + \sum_r (n_{\text{f}}\bar{s}_r) - \sum_p (n_{\text{f}}\bar{s}_p) + \dot{S}_{gen}$$  

Which for steady state assume $\frac{dS}{dt} = 0$, then the last equation become:

$$\frac{\dot{Q}_1}{T_1} + \dot{S}_{gen} = \left[ \left( n_{\text{f}}\bar{s}_{\text{H}_2\text{O},b}\right)_b + \left( n_{\text{f}}\bar{s}_1 + n_{\text{f}}\bar{s}_b \right)_b \right]$$  

Dividing the last equation by $n_{\text{f}}\text{H}_2$ yields:

$$\frac{\dot{q}_1}{T_1} + \dot{S}_{gen} = \left( \bar{s}_{\text{H}_2\text{O},b}\right)_b + \left( \bar{s}_1 + \bar{s}_b \right)_b = \bar{s}_{RF}$$  

Noting that the reaction inside an ideal fuel cell is internally reversible and the heat transfer crosses the system boundary at temperature $T_{\text{FC}}$, $q_{\text{FC}}$ can be calculated from (8) as:

$$\bar{q}_{\text{FC}} = T_{\text{FC}} \bar{s}_{RF}$$

The heat of fuel cell ($q_{\text{FC}}$) can be calculated from (9) with $T_{\text{FC}}$ and then substituted into (5) to find $W_{\text{FC, elec}}$.

2.4. Gibbs free energy calculation

The inputs and outputs of the basic fuel cell are shown in Figure 3.2. The electrical power and energy output are easily calculated from the well known Formulas, [9,10,11].

$$\text{Power} = V.I$$
$$\text{Energy} = \text{Power}.t = V.I.t$$

To analyze the chemical energy changes throughout the chemical process involved in the operation of a fuel cell, one must be aware of and understand “Gibbs free energy”. This is defined as the “energy available” to do external work, neglecting any work done by changes in pressure and/or volume, [12].

For chemical energy the point of zero energy can be define as almost anywhere. “Gibbs free energy of formation”, is used when this convention is used. Therefore, the Gibbs free energy of formation is zero for the input state, thus simplifying calculations and creating a standard. Then $G_f$ of both $\text{O}_2$ and $\text{H}_2$ is
zero, a useful result when dealing with a hydrogen oxygen fuel cell. Where \( \Delta G_f \) refers to the difference in Gibbs free energy of formation between the inputs and the outputs, and is therefore a specific measure of the energy released by the reaction.\[13, 14]\.

\[ \Delta G_f = \Delta G_{f,P} - \Delta G_{f,R} \]  

(11)

Usually this quantity is per a mole of chemical. So the chemical energy released during a nominal fuel cell reaction (1). The Gibbs function of a system is defined in terms of the entropy and the enthalpy:

\[ G = H - T \cdot S \]  

(12)

Here, \( T \) is the temperature of the system and \( S \) is the entropy, or disorder, of the system. \( H \) is the enthalpy of the system.\[15,16]\.

Similarly, the molar Gibbs energy of formation, the molar enthalpy of formation, and the molar entropy are connected by the equation,\[10,11]\.

\[ \tilde{g}_f = \tilde{H}_f - T \cdot \tilde{S}_f \]  

(13)

In this case, the change in energy that is important. In addition, the temperature in a fuel cell is constant. So the change in the molar Gibbs energy of formation is:\[10,14,17].

\[ \Delta \tilde{g}_f = \Delta \tilde{H}_f - T \cdot \Delta \tilde{S}_f \]  

(14)

The value of \( \Delta \tilde{H}_f \) is the difference between \( \tilde{H}_f \) of the products and \( \tilde{H}_f \) of the reactants. So for the reaction (1), the product is one mole of \( (H_2O) \) and the reactants are one mole of \( H_2 \), and half a mole of \( O_2 \). Thus:

\[ \Delta \tilde{H}_f = \tilde{H}_{f,H_2O} - \tilde{H}_{f,H_2} + \frac{1}{2} \tilde{H}_{f,O_2} \]  

(15)

Similarly, \( \Delta \tilde{S}_f \) is the difference between entropy of the products and reactants so that:

\[ \Delta \tilde{S}_f = \tilde{S}_{f,H_2O} - \tilde{S}_{f,H_2} + \frac{1}{2} \tilde{S}_{f,O_2} \]  

(16)

The values of \( \tilde{S}_f \) vary with temperature and pressure according to the equations given below. In these equations, the subscript to \( h \) and \( s \) is the temperature, and \( \tilde{C}_p \) is the molar heat capacity at constant pressure. The standard temperature is 298.15 K, which is necessary as an integration limit. The “\( T \)” subscript to the enthalpy, \( \tilde{H}_f \), means the enthalpy at temperature \( T \).

The enthalpies of stable elements and compounds, \( \tilde{H}_{f,T,P}^{\text{298}} \). In the more general case the enthalpies of the reactants and products must be evaluated at a state \((T, P)\) different from the reference state, \((T_0, P_0)\)\[15,16,17].

Symbolically, this requires that:

\[ \tilde{H}_{f,T,P} = \tilde{H}_{f,T,P}^{\text{298}} + (\tilde{H}_{f,T,P}^{\text{298}} - \tilde{H}_{f,T,P}^{\text{298}})_{T,P} \]  

(18)

Where represent the difference in enthalpy between any given state and the enthalpy of ideal gas at 298.15 K, 0.1MPa (1atm), Table I,\[15,17].

In this case, the enthalpy is a function of the temperature only, and can found by use of an equation of as:

\[ \tilde{H}_{f,T,P} = \tilde{H}_f^{298} + \int_{298}^{T} \tilde{C}_p dT \]  

(19)

or from tabulated value of enthalpy as function of temperature (which assumes ideal gas behavior),\[17,18].

Similarly, the molar entropy, \( \tilde{S}_f \) at temperature \((T)\) is given by:

\[ \tilde{S}_f = \tilde{S}_f^{298} + \int_{298}^{T} \frac{\tilde{C}_p}{T} dT \]  

(20)

The molar heat capacity at constant pressure, \( \tilde{C}_p \). Over a range of temperatures, \( \tilde{C}_p \) is not constant temperature but can be described by “Nevertheless”, empirically, the molar heat capacity at constant pressure of a species, as a function of \( T \), can be expressed as:

\[ \tilde{C}_p = \tilde{M} \left[ A + B T + C T^2 + D T^3 \right] \]  

(21)

Where \( \tilde{M} \) is the molecular mass and A, B, C and D are empirical constants, These are listed in the Table 1,\[17].

By substituting into (19) and (20), giving functions that can be readily integrated and thus evaluated at any temperature \((T)\). This is done to derive values for \( \tilde{H}_{f,T,P} \) and \( \tilde{S}_f \) chemical reaction (reactants and products), then drive values \( \Delta \tilde{g}_f \) and \( \Delta \tilde{S}_f \), which are finally substituted into (14), to giving
the change in molar Gibbs energy of formation \( \Delta_{gT} \), [19,20]. Then for hydrogen fuel cells reaction the change in the Gibbs free energy of formation per mole becomes:

\[
\Delta_{g} = (\overline{g}_{H_2O})_{H_2O} - (\overline{g}_{H_2})_{H_2} - \frac{1}{2}(\overline{g}_{O_2})_{O_2} \]

(22)

Where \( \Delta_{gT} \), a change for different molecular states of the materials in the fuel cell and at different fuel cell temperatures.

If the material changes state then \( \Delta_{gT} \) must change accordingly. The states for the reactants and products in the fuel cell do not change. These are shown: \( H_2 \) (gas), \( O_2 \) (gas), \( H_2O \) (Liquid). [20]. Note that the water product is expected to be a liquid and gas mixture but the gas content is small enough to be neglected, [21].

2.5. Reversible work and efficiency of fuel cell

The electrical work done by the fuel cell in moving two electrons around the circuit is given by

\[ W_{max} = \Delta_{gT} = \text{nFE} \]

(23)

Where, E is the voltage of the fuel cell.[21] If the process is reversible for hydrogen reaction, then all this Gibbs free energy is converted into electrical energy, then the Gibbs free energy can be used to find the open circuit voltage of the fuel cell. Since the process is reversible then the electrical work done will be equal to the Gibbs free energy released, [4,10, 14]

Then:

\[ W_{rev} = W_{max} = \Delta_{gT} = \frac{\Delta_{gT}}{2} \text{nFE} \]

(24)

This fundamental equation gives the electromotive force (EMF) or reversible open circuit voltage of the hydrogen fuel cell [12, 13, 35].

The reversible electrical efficiency of a fuel cell is defined, by (25) assuming that all reactants and products are in their standard state: [18, 24, 36]

\[ \eta_{rev} = \frac{\Delta_{gT}}{\Delta_{H}T} \times 100 \]

(25)

Typical energy converters that involve high temperatures are limited by the Carnot efficiency, which is given as:

\[ \text{Carnot Efficiency} = 1 - \frac{T_L}{T_H} \]

(26)

Where \( T_L \) and \( T_H \) are the temperature extremes of the cycle. Since a fuel cell is not a heat engine, it is not bound by the Carnot efficiency.

2.6. Using C++ computer program for the modeling of PEMFC

The computer program (C language) that is presented in Figure.3, is designed to be effective in calculation of heat, Gibbs free energy, reversible potential, electrical work and efficiency of fuel cells. There are many types of fuel used in this program, therefore, it can used for all types of fuel cells.

The first step for this program is the determination of the specific heat under constant pressures for the most common chemical elements, where the program achieve the normal integration
process for the general specific temperature equation under constant pressure, Equ (21), the program reads the values of the constants that in equation; in addition to the enthalpy, entropy, and the Gibbs free energy at temperature of \(25^\circ\text{C}\) (298K). Where these values are considered as constants read from the tables of ideal gas properties at temperature of \(25^\circ\text{C}\), which are present in Table (1), the program also depends on the type of the used fuel, which determines the chemical formula of the fuel cell; there are three options to determine the chemical formula; the first is in the case of hydrogen fuel or one of the hydrocarbons. Where the general reaction equation that used in this program for 1mol is as Equ (27), [22].

\[
\begin{align*}
C_xH_y + \alpha(x + \frac{y}{4})(O_2 + 3.76N_2) &\rightarrow \ \\
xCO_2 + \frac{y}{2}H_2O + (\alpha - 1)(x + \frac{y}{4})O_2 &+ \\
3.76\alpha(x + \frac{y}{4})N_2
\end{align*}
\]

Where \(\alpha\) corresponds to the stoichiometric amount of air (a percent theoretical air \(\alpha \geq 100\%\), [11].

For the PEM fuel cell the hydrogen is used as a fuel, in this case the carbon atoms are not present (\(x = 0\)).

Figure (3). The flow chart of the calculation procedure of the PEM moled

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3. RESULTS AND DISCUSSION

The above thermo-chemical model is solved by creating (C++) computer program, all the calculations of this cell depend on the range of operating temperatures. Which range between 50 and 100°C (323-373 K), this range is less than the evaporation temperature of water, so the water producing from the reaction of hydrogen and pure oxygen in this cell is in the liquid state. It assumed that the water state at 100°C is the liquid state.

The calculation of the producing energy from the fuel cell depends on the entropy and enthalpy of reaction, where the enthalpy represents the heat energy produced from the reaction of the fuel cell. Figure 4, shows the enthalpy is constantly increasing, and so the curve is a straight line. The negative sign indicates that the fuel cells produce heat energy.

The relation between the producing heat energy (change in enthalpy) and the operating temperature for the PEMFC cell is a proportional relation. Whereas the relation of entropy with the operating temperature is similar to that of the enthalpy, where the entropy increases also with increasing the operating temperatures as shown in Figure 5, the values of entropy are in KJ/Kmol, and the temperatures are in Kelvin. The best operating temperature of this cell is the maximum temperature where the cell produces the highest energy at this temperature.

The results involve some electrical calculations such as the reversible potential. It depends on the Gibbs free energy and the type of the reaction.

Figure (4). Enthalpy of Reaction Behavior of PEMFC

Figure (5). Entropy of Reaction Behavior of PEMFC

The Gibbs free energy relates both entropy and enthalpy, and is a function of the operating temperature, where its behavior is somewhat different from that previously mentioned. This is because Gibbs energy decreases gradually with temperature as shown in Figure 6. In other words, Gibbs energy is at its highest value at the lowest operating temperature, and decreases gradually with increasing the temperature.

The results involve some electrical calculations such as the reversible potential. It depends on the Gibbs free energy and the type of the reaction.

Figure (6). Gibbs Free Energy Behavior of PEMFC

Figure 7 shows the gradual decrease of the
reversible potential, it is irregular and did not represent by a straight line. Where it shows a little decrease at a specific range of temperature, and is constant at another range of operating temperature, this means that the curve of the reversible potential oscillates between decrease and constant and vice versa.

Figure (7). Effect Operating Temperature on PEMFC Reversible Potential

The heat output (required heat) from the fuel cell increases with increasing the operating temperature, it similar to the behavior of enthalpy and entropy as shown in Figure. 8.

Figure (8). Effect Operating Temperature on Heat Output from PEMFC

The behavior of the reversible electrical work with the operating temperature of the fuel cell (reversible electrical work or the maximum work) is similar to the behavior of Gibbs free energy with different sign as shown in Figure. 9, where the highest work dose recorded at the lowest operating temperature of this cell.

Figure (9). PEMFC Electrical Work Behavior

The efficiency of the fuel cell is not limited by Carnot efficiency, which is considered the highest efficiency of the heat engines, so it the efficiency of the ideal engine.

There is a break between (31% - 72%) in the Y-axis that represents the efficiency as shown in Figure. 10. Where the axis divided into two parts, the first part represents Carnot’s efficiency, while the second part of the Y-axis represents the efficiency of the fuel cell (PEMFC). The efficiency of this type of the fuel cells is much higher than the ideal Carnot’s efficiency.

Figure (10). Comparisons between PEMFC and Carnot Efficiencies
4. CONCLUSIONS

Fuel cells directly convert the chemical energy in hydrogen to electricity, with pure water and potentially useful heat as the only byproducts. Hydrogen-powered fuel cells with free pollution, the PEM fuel cell can generate electricity at efficiencies up to 80% depends on temperature at which it operates.

The results show that the values of enthalpy and entropy of the fuel cell chemical reaction increase with the operating temperature increase, where their maximum values of are at the maximum operating temperature of the PEM fuel cell.

The change in the enthalpy and entropy values with operating temperature range are affects positively on the heat produced by the fuel cell, while the reversible work, electric potential and Gibbs free energy are changes inversely with the operating temperature, where the maximum values of these variables are at the PEM fuel cell operation temperature.

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