Modelling and Examining Open Circuit Voltage for PEM Fuel Cells

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Abstract - Fuel cells are ultimately one of the best promising devices that convert the potential energy in the reactants into the electrical energy after steps of sequential electrochemical processes with no hazards up on the environment. This paper presents comprehensive derivation and understanding of the electrochemical reaction for Polymer Electrolyte Membrane Fuel Cell (PEMFC) in terms of parameters and surrounding impacts, starting from the initial phenomena of splitting hydrogen molecule to the final mathematical model of open circuit voltage for PEMFC. The open circuit voltage has been derived and modelled for two operational conditions, when the output water from fuel cell is in liquid state, and when the output water is in state of vapour. Examining and analysing OCV under different operational temperature and pressure. Using pure oxygen instead of air has very small impact on increasing the OCV by 1% under same operational temperature and pressure; hence the use of pure oxygen is no longer defensible motive as this will increase the weight and cost of the system.

Keywords: PEM Fuel Cell, Open Circuit Voltage, Mathematical Modelling, Electrochemical Reaction.

1. Introduction

The first use of fuel cells as an auxiliary power source device was in 1960s in the Gemini space flights program directed by NASA. The polymer electrolyte membrane fuel cells (PEMFCs), also known as proton exchange membrane fuel cells was developed and deployed in this program [1].

Fuel cell is a device which directly converts the energy in the fuel and reactant into electricity. The efficiency of energy conversion-production for FC is comparatively higher than internal combustion engines because there is no intermediate thermal conversion process similar to the internal combustion engine or gas turbine [2].

Fuel cells can be considered as one of the finest reliable energy sources in producing safe and environmentally clean energy with low cost effectively [3]. More interests and concerns are turned toward PEM fuel cells to be more developed than other types of fuel cell, as PEM fuel cells have: high power density, high conversion efficiency, solid polymer electrolyte, long operational life hours, low corrosion, low weight and compact size which make them suitable for mobile and stationary applications, and zero pollution emissions. Add to characteristics, low these operational temperatures (30-100°C) providing faster dynamical response in comparing to other types of fuel cells, which enables fast start-up and make them suitable for mobile and stationary applications [4], [5], [6], [7], [8].

Comparing with conventional batteries, fuel cells produce higher energy density. Also in vehicle application, the time consumed in recharging the battery is longer than the time spent in refuelling the storage tank by hydrogen [4]. Therefore major automotive companies such as Ford, Nissan, Daimeler Chrysler are developing electric vehicles based on PEMFC.

In general, the output voltage of the PEMFC depends on the operational pressure, temperature, reactants concentration at the electrodes, mass flow rate of the reactants, and operational losses of the FC, also the voltage of the FC drops when the current drawn from it [2].

2. Electrochemical Reaction of PEMFC

In a chemical laboratory, a combusting of hydrogen and oxygen to produce water requires energy equal to 432 KJ/mole in order to break the covalent bond between the two atoms of hydrogen molecule. This activation energy is required to start the reaction and is usually provided by spark or flame. After the reaction is initiated, the initial energy released will provide the necessary energy to continue breaking others hydrogen atoms. A catalyst could be used to reduce the essential activation energy and also to allow the reaction to proceed without the necessity to the initial energy provided by flame or spark [9].

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At the anode side of the PEM fuel cell, the hydrogen will be ionized in the existence of platinum catalyst to release electrons and H⁺ ions in a process called oxidization, each molecule of hydrogen H₂ will produce two free electrons and two positive ions. The existence of platinum catalyst is important to accelerate the reaction (i.e. separation process) and reducing the activation energy. The mass of platinum in the electrode is not going to be depleted with time because it does not contribute in the reaction [1].

The ions and electrons will not produce immediately at the first instant of contact between H_2 and catalytic electrode, the process is more complicated and it could pass though many stages. H_2 molecule will be adsorbed on the surface of electrode, the energy of the interaction between the hydrogen and platinum surface will contribute in breaking the bonds between the hydrogen atoms. Hence, the hydrogen molecule will be separated into two adsorbed hydrogen atoms (H_{ad}), and then each adsorbed atom will be dissociated into one electron and one ion [9].

$$H_2 \rightarrow 2H_{ad}$$

 $2H_{ad} \rightarrow 2H^+ + 2e^-$

The electrolyte membrane will conduct ions to the cathode electrode, but it will not allow the electrons to be conducted through, because it has high resistivity towards electrons. Therefore, the electrons will flow as an electrical current through the electrical circuit via the connections of bipolar plates [10]. At the cathode side, the oxygen will react with the electrons and ions in the existence of platinum catalyst to produce water in a process called a reduction [1]. The electrochemical reaction of the PEM fuel cell is associated with releasing energy in the form of heat. The equations below represent the form of the reaction.

$$H_2 \rightarrow 2H^+ + 2e^{-1}$$
 (at anode side)
 $2H^+ + 2e^{-1} + \frac{1}{2} O_2 \rightarrow H_2O$ (at cathode side)

Fuel cell can be considered as a complex system subjects to different laws and theories of thermodynamics, physics, and chemistry. Deriving and modelling the electrochemical reaction involved in the operation of the fuel cell is critical.

Enthalpy H is a partial measure for the internal energy of the system, and ΔH is the change in enthalpy of the system. In a chemical reaction, the change in the enthalpy of formation is equal to the total energy released, this change is equal to the total amounts of the enthalpy of formation for the products minus the total amounts of the enthalpy formation for the reactants [11].

$$\Delta H = \sum (\Delta H)_{\text{products}} - \sum (\Delta H)_{\text{reactants}}$$

The changes in the enthalpy of formation ΔH are equivalent to the higher heating values (HHV) of the fuels. Values of changes in the enthalpy of formation and Gibbs free energy of compounds and elements are given in Table 1 for the oxidation of several fuels at 25°C and 1atm. ΔH is negative for any exothermic reaction and positive for any endothermic reaction [12] & [13].

Table 1: Values of enthalpy of formation and Gibbs free energy of compounds and elements, at 25°C & 1atm.

Compounds or Elements	State	ΔH, KJ/mol	ΔH, KJ/gm	∆G, KJ/mol	ΔG, KJ/gm
O_2	Gas	0	0	0	0
N ₂	Gas	0	0	0	0
H_2	Gas	0	0	0	0
СО	Gas	-110.54	-3.947	-137.5	-4.9108
CO ₂	Gas	-393.52	-8.944	-395.0	-8.977
H ₂ O	Liquid	-285.84	-15.880	-237.0	-13.167
H ₂ O	Vapour	-241.83	-13.435	-228.0	-12.667
CH ₄	Gas	-74.87	4.679	-50.8	-3.175
H ⁺	-	0	0	0	0
OH.	-	-230.0	-13.530	-157.0	-9.236

From Table 1, lower heating value (LHV) of water is equal to $\Delta H = -241.83 \text{ kJ.mol}^{-1}$ (for vapour state), and higher heating value (HHV) is equal to $\Delta H = -285.84 \text{ kJ.mol}^{-1}$ (for liquid state). The difference between these two values (44.01 kJ.mol⁻¹) represents the molar enthalpy of vaporization of water [10].

If the thermodynamic process is ideally been presumed to go through a sequence of states that are infinitesimally close to equilibrium, hence the system at any instant remains in equilibrium (no dissipation or loss in the energy of the system), and in this case the process is typically reversible [14]. In order to simplify deriving the necessary equations of the

electrochemical reaction for PEM fuel cell, the process will be initially considered to be reversible.

Enthalpy of formation (H) is equal to the sum of the Gibbs free energy of the formation (G) and the released heat (Q), [12].

$$H = G + Q \tag{1}$$

$$\Delta H = \Delta G + \Delta Q \tag{2}$$

The change in this Gibbs free energy of formation ΔG_f is equal to the sum of the Gibbs free energy formation for the products minus the sum of the Gibbs free energy formation for reactants.

$$\Delta G_f = \sum (\Delta G_f)_{\text{products}} - \sum (\Delta G_f)_{\text{reactants}}$$

Assuming theoretically, that all the energy in the enthalpy of formation is going to be converted into electrical energy (W_e), thus none of energy in the enthalpy were converted into any other form of energy ($\Delta H = \Delta G = W_e$, and $\Delta Q = 0$). But practically, not all the energy in the enthalpy of formation is converted into electrical energy, part of this energy is released as thermal energy; the remaining of this energy could be converted into electrical energy.

According to the second law of thermodynamics, when heat is transferred to or from the system, temperature is associated with the transported heat [11]. The amount of released thermal energy is given as $(Q = \int T.dS)$. Where, T is the absolute temperature, and S is the entropy property of thermodynamic system. If fuel cell is initially considered as an isothermal process dT = 0, then,

$$Q = T.\int dS = T.\Delta S \tag{3}$$

By differentiating equation (1) and rearranging, the amount of electrical energy produced is given as,

$$dH = dG + T.dS \tag{4}$$

$$W_e \le \Delta H - T.\Delta S$$
, or $W_e \le \Delta G$ (5)

For a reversible reaction (i.e. no losses in fuel cell reaction), the electrical energy is equal to $(\Delta H-T.\Delta S)$. While for irreversible reaction, the electrical energy is less than $(\Delta H-T.\Delta S)$.

Based on the first law of thermodynamics "the increment in the internal energy of a closed system is equal to the heat provided to the system plus the work done on the system" [11].

$$U_2 - U_1 = Q_{12} + W_{12} \tag{6}$$

Where, Q and W refer to the heat and work respectively, U_1 and U_2 represent the internal energy of the system were the process moves between states 1 and 2. For reversible system passes through continuous series of equilibrium in their initial and final states. Then,

$$U_2 - U_1 = Q + W (7)$$

$$dU = dQ + dW \tag{8}$$

When work is going to be delivered to the surroundings, then (dW = -P.dV) [11]. Therefore,

$$dU = dQ - P.dV (9)$$

By substitute (dQ = T.dS) from above in equation (9),

$$dU = T.dS - P.dV \tag{10}$$

Enthalpy of the formation is defined according to the first law of thermodynamics, as given below [13],

$$H = U + P.V \tag{11}$$

$$dH = dU + P.dV + V.dP \tag{12}$$

By substituting equation (10) in equation (12), yields with,

$$dH = T.dS - P.dV + P.dV + V.dP$$

$$dH = T.dS + V.dP$$
(13)

By substitute equation (13) in equation (4) and rearranging, yields with,

$$dG = T.dS + V.dP - T.dS$$

$$dG = V.dP$$
(14)

The ideal gas equation is,

$$P.V = n.R.T \tag{15}$$

Where, P is the pressure of gas (Bar or Pascal), V is the volume of gas (m^3), T is absolute temperature in Kelvin, R is Universal Gas Constant (R = 8.31441 KJ/Kmol·K), and n represents number of moles and n is equal to mass (gm) divided by molar mass(gm/mol), [14]. By substituting equation (15) in equation (14) and for n = 1, yields with,

$$dG = \frac{R.T}{P}.dP \tag{16}$$

If G_1 is the value of Gibbs free energy at initial pressure P_1 , and G_2 is the value of Gibbs free energy at final pressure P_2 . For an isothermal process and by integrating the two sides of equation (16), at constant temperature when the pressure of the system changes from P_1 to P_2 , then the change in molar Gibbs free energy will be,

$$\int_{G_1}^{G_2} dG = \int_{P_1}^{P_2} \frac{RT}{P} dP = G_2 - G_1 = RT \ln \frac{P_2}{P_1}$$

$$G_2 = G_1 + RT \ln \frac{P_2}{P_1} \tag{17}$$

If state 1 is referenced to pressure P° (1atm), the change Gibbs free energy G at certain pressure P is,

$$\int_{G}^{G} dG = \int_{P}^{P} \frac{RT}{P} dP$$

$$G = G^{\circ} + RT \ln \frac{P}{P^{\circ}} \tag{18}$$

Gibbs free energy of formation is function to the changes in temperature and pressure of the reactants. For hydrogen fuel cell, two electrons are passing over the external circuit for each one molecule of hydrogen consumed. If (-e) is the charge of one electron; then the total charge of released electrons for one mole of hydrogen molecules been used in the reaction is equal to $(-2 N_A$. e = -2 F Coulombs/mol). Where, N_A is Avogadro's number $(6.022 * 10^{23})$ and the charge of one electron is $(1.601 * 10^{-19} \text{ coulombs})$. Charge of one mole of electrons is known as Faraday's constant F.

$$F = N_A$$
 . e = 6.022 * 10^{23} * 1.601 * 10^{-19} = 96485 Coulombs/mol.

If E is the open circuit voltage of the fuel cell, then the electrical work done in driving these electrons round the electrical circuit is

Electrical work done = Charge * Voltage = $-2 N_A$. e. E = -2 F. E (joules).

Ideally, if there is no losses associated the fuel cell reaction, the electrical work done to drive these electrons round the external circuit will be equal to the released Gibbs free energy, then $(\Delta G = -2 F.E)$.

$$E = \frac{-\Delta G}{2F} \tag{19}$$

By substituting values of Gibbs free energy from Table 1 in equation (19) to determine the value of ideal open circuit voltage E° of hydrogen fuel cell at standard pressure 1atm, and temperature 25°C.

For case of output water from fuel cell is in liquid state (HHV), $\Delta G = -237$ KJ/mol, and $E^{\circ} = 1.228$ V. And for case of water in vapour state (LHV), $\Delta G = -228$ KJ/mol, and $E^{\circ} = 1.182$ V.

Dividing equations (17) and (18) by -2F, and rearranging, yields with,

$$\frac{-\Delta G_2}{2F} = \frac{-\Delta G_1}{2F} - \frac{RT}{2F} \ln \frac{P_2}{P_1}$$

$$E_2 = E_1 - \frac{RT}{2F} \ln \frac{P_2}{P_1} \tag{20}$$

$$E = E^{\circ} - \frac{RT}{2F} \ln \frac{P}{P^{\circ}}$$
 (21)

Where, E_1 and E_2 are the open circuit voltage for fuel cell at the initial and final states respectively, and P_1 and P_2 are the pressure or partial pressure of the system at these states.

3. Effect of Pressure and Temperature on OCV for PEM Fuel Cell

For an ideal gas; the gas activity is equal to the ratio of the pressure or partial pressure of the gas over the standard pressure [10].

activity,
$$a_i = \frac{P_i}{P^{\circ}}$$
 (22)

Where, (P_i) is the pressure or partial pressure of gas (i), and (P^o) is standard pressure (1atm). Fuel cells are in general gas reactor devices; therefore the activity is proportional to partial pressure of the gas. In the case of hydrogen fuel cell reaction, the water produced from fuel cells could be either in state of vapour or liquid or a mixture of both, which makes the calculation a little bit difficult. Based on equation (22), the activities of reactants and products can be determined as,

$$a_{H_2} = \frac{P_{H_2}}{P^{\circ}} = P_{H_2} \tag{23}$$

$$a_{O_2} = \left(\frac{P_{O_2}}{P^{\circ}}\right)^{1/2} = \left(P_{O_2}\right)^{1/2} \tag{24}$$

$$a_{H_2O} = \frac{P_{H_2O}}{P_{H_2O}^{\circ}} \tag{25}$$

Where, P_{H_2O} is the vapour pressure of water in the mixture, and $P^o_{H_2O}$ is vapour pressure of pure water (e.g. distilled water) at specific temperature, and it could be determined from the vapour tables. If we assume that at high temperature, the water vapour behaves like an ideal gas, then $(P^o_{H_2O} = P^\circ)$ and equation (25) becomes,

$$a_{H_2O} = \frac{P_{H_2O}}{P^O} = P_{H_2O} \text{ (Vapour state)}$$
 (26)

If the chemical reaction occurs at constant pressure and temperature, the reactant gases A and B are reacted to form the products M and N [15].

$$aA + bB \rightarrow mM + nN$$

Where, a, b, m, and n are the stoichiometeric coefficients. Then, the change in Gibbs free energy is,

$$\Delta G = mG_M + nG_N - aG_A - bG_B$$

And the chemical reaction of the fuel cell could be expressed by substituting equation (18) in each of the four terms, yields with,

$$\Delta G = m \left[G_{M}^{\circ} + RT \ln \left(\frac{P_{M}}{P^{\circ}} \right) \right] + n \left[G_{N}^{\circ} + RT \ln \left(\frac{P_{N}}{P^{\circ}} \right) \right]$$
$$-a \left[G_{A}^{\circ} + RT \ln \left(\frac{P_{A}}{P^{\circ}} \right) \right] - b \left[G_{B}^{\circ} + RT \ln \left(\frac{P_{B}}{P^{\circ}} \right) \right]$$

The change in standard Gibbs free energy ΔG° can be expressed as.

$$\Delta G^{\circ} = mG_{M}^{\circ} + nG_{N}^{\circ} - aG_{A}^{\circ} - bG_{R}^{\circ}$$

$$\Delta G = \Delta G^{\circ} + RT. \ln \frac{P_M^m . P_N^n}{P_A^a . P_B^b}$$
 (27)

For hydrogen fuel cell reaction, $H_2 + \frac{1}{2} O_2 \rightarrow H_2 O_2$ then

$$\Delta G = \Delta G^{\circ} + RT. \ln \frac{P_{H_2O}}{P_{H_2} \left(P_{O_2}\right)^{1/2}}$$

$$\Delta G = \Delta G^{\circ} - RT. \ln \frac{P_{H_2} \cdot (P_{O_2})^{1/2}}{P_{H_2} O}$$
 (28)

By substituting equation (19) in equation (28), yields with,

$$E = E^{\circ} + \frac{RT}{2F} \cdot \ln \frac{P_{H_2} \cdot (P_{O_2})^{1/2}}{P_{H_2O}}$$
 (Vapor state) (29)

By substituting equations (23), (24), and (26) in equation (29), yields with,

$$E = E^{\circ} + \frac{RT}{2F} \cdot \ln \frac{{}^{a}H_{2} \cdot {}^{a}O_{2}}{{}^{a}H_{2}O}$$
 (Vapor state) (30)

In order to determine the impact temperature changes upon the open circuit voltage of the fuel cell, ΔE_t represents the change in open circuit voltage as a result of temperature changes. Then,

$$E_{OC} = E^{\circ} + \Delta E_t + \frac{RT}{2F} \ln \frac{a_{H_2} \cdot a_{O_2}}{a_{H_2O}} \text{ (Vapor state) (31)}$$

From above equitations (2 & 3), $\Delta Q^{\circ} = \Delta H^{\circ} - \Delta G^{\circ} = T^{\circ}.\Delta S^{\circ}$, using the values of ΔH & ΔG given for water in Table 1, at 25°C and 1atm.

$$\Delta S^{\circ} = \frac{\Delta H^{\circ} - \Delta G^{\circ}}{T^{\circ}}$$

$$= \frac{241.83 - 228}{298.15} = 46.387 \text{ J/mol.K} \qquad \text{(Vapor state)}$$

$$\Delta S^{\circ} = \frac{285.84 - 237}{298.15} = 163.8$$
 J/mol.K (Liquid state)

At a certain temperature, the amount of heat energy produced form the reaction is

$$\Delta Q = -2.F.\Delta E_t = \Delta T. \ \Delta S^{\circ} = (T - T^{\circ}).\Delta S^{\circ}$$
$$= (T - 298.15). \ \Delta S^{\circ}$$

$$\Delta E_t = \frac{(T - 298.15) * 46.387}{-2 F}$$
 (Vapor state) (32)

$$\Delta E_t = \frac{(T - 298.15) * 163.8}{-2.F}$$
 (Liquid state) (33)

Where, ($T^{\circ} = 25^{\circ}\text{C}+273.15 = 298.15 \text{ kelvin}$), and (F = 96485Columbs/mol). By substituting equation (32) in equation (31) and for value of $E^{\circ} = 1.182 \text{ V}$ (for LHV, vapour state), yields with,

$$E_{oc} = 1.182 - 0.24 * 10^{-3}.(T - 298.15) +$$

$$4.3086 * 10^{-5} * T \ln \frac{a_{H_2}.a_{O_2}}{a_{H_2}O} \text{ (Vapor state)}$$
(34)

As the product of the fuel cell is almost in liquid state, then the activities of water could be assumed to be approximately equal to unity $(a_{H2O} = 1)$ [10]. Similarly, by substituting equation (33) in equation (31), and for value of $E^{\circ} = 1.228V$ (for HHV, liquid

$$E_{oc} = 1.228 - 0.85 * 10^{-3}.(T - 298.15) + 4.3086 * 10^{-5} * T.\ln(a_{H_2}.a_{O_2}) \text{ (Liquid)}$$
(35)

$$E_{oc} = 1.228 - 0.85 * 10^{-3} . (T - 298.15) +$$

$$4.3086 * 10^{-5} * T. \ln \left(P_{H_2} . (P_{O_2})^{1/2} \right)$$
(36)

4. Results and Discussions

state), yields with,

Using equation (36) to determine the values of open circuit voltages, when temperature increases

from 25°C to 90°C and pressure of both fuel and reactant (i.e. reactant of oxygen or air) increases from 1.0 to 6.0 bar at step of 1.0 bar. Figure 1 shows the values of open circuit voltages for Hydrogen-Oxygen fuel cell, while Figure 2 shows the values of open circuit voltages for Hydrogen-Air fuel cell. A rapid increase in pressure form 1 bar to 6 bar will lead to increase the open circuit voltages by gain of 32mv to 42mv. But when the pressure of both fuel and reactant increases gradually from 1.0 bar to 6.0 bar at step of 0.1 bar, the gain in the output voltages are very small between 1.5mv to 1.75mv as depicted in figures 3 and 4. Therefore, it could be concluded that a gradual reduction in the pressure of fuel cell will have minor impact on the OCV of the fuel cell, hence a rapid drop in the pressure must be avoided in order to avoid high drop in the output voltage.

It is clear that increasing temperature from 30°C to 90°C will lead to decrease the open circuit voltages by about 4% as depicted in Figures 3 & 4. Moreover, using pure oxygen to feed the fuel cells has an impact of 1% gain in the OCV comparing with using fresh air for fuel cell operating at same temperature and pressure, as depicted in Figures 1 & 2. Hence, there is no point of using pure oxygen to feed the fuel cell, as this will increase the weight and cost of the system.

However, the voltage drops would be increased when many cells are connected in series and useful current drawn from it. Also when many cells are connected in series, the manifold channel will provide resistance against the flow of reactants causing difficulties in transporting sufficient amount of reactants.

5. Conclusion

A comprehensive derivation and understanding of the electrochemical reaction for PEM fuel cell in terms of parameters and surrounding impacts, also examining and analysing OCV under different operational temperature and pressure are presented and discussed in this work. Gradual reduction in the pressure of fuel cell will have minor impact on the OCV of the fuel cell, hence a rapid drop in the pressures must be avoided in order to avoid high drop in the output voltage.

Using pure oxygen instead of air has very small impact on increasing the OCV by 1% under same operational temperature and pressure. Hence, the use of pure oxygen is no longer justifiable motive as this will increase the weight and cost of the system.

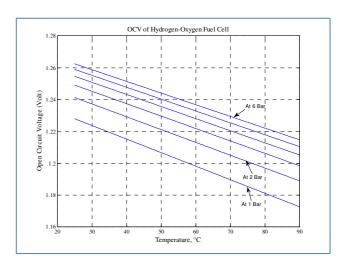


Figure 1: Open circuit voltages of Hydrogen-Oxygen fuel cell when pressures change in step of 1 bar.

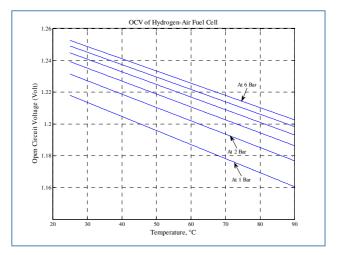


Figure 2: Open circuit voltages of Hydrogen-Air fuel cell when pressures change in step of 1 bar.

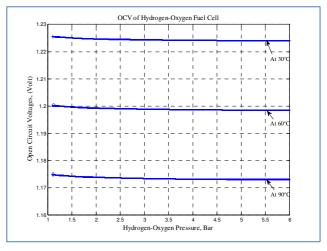


Figure 3: Open circuit voltages of Hydrogen-Oxygen fuel cell when pressures change in step of 0.1 bar

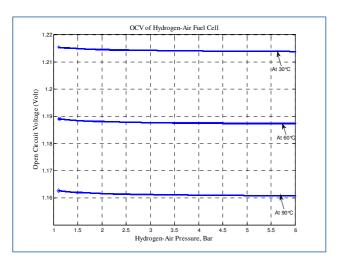


Figure 4: Open circuit voltages of Hydrogen-Air fuel cell when pressures change in step of 0.1 bar

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