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Determination of thermodynamic parameters and design of a hydrogen fuel cell level pilot plant[1]

Determinación de parámetros termodinámicos y de diseño de una celda combustible de hidrógeno a nivel de planta piloto.

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Abstract **- The clean alternative energy production is a challenge facing society, therefore, through the analysis and design made a prototype production cell hydrogen proton-type pilot plant level (PEM), operating in potentiostatic conditions 1V, 1.23 V and 5 V and galvanostatic conditions of 30 mA, 40 mA, 50 mA, 70 mA, 80 mA y 90 mA for 10 minutes, using KOH as a catalyst and four steel electrodes preactivated with 3 mm separation between them (73 cm2 surface), It was permitted to obtain the thermodynamic parameters, such as the enthalpy of the reaction, the Gibbs free energy and entropy; besides it was stablished optimal potential experimental work and energy efficiency when a Nafion 117 membrane on (surface 36 cm2) was used and in the absence of the same.**

Key Words **- Thermodynamic properties, fuel cell (electrochemical reactor), Nafion 117 (Nafion 117), potentiostatic, galvanostatic (galvanostatic).**

Resumen **- La producción limpia de energías alternativas, es un reto que afronta la sociedad, por ello, a través del análisis y diseño realizado a un prototipo de celda de producción de hidrógeno tipo protónico a nivel de planta piloto (PEM), operando en condiciones potenciostáticas de 1 V, 1,23 V, y 5 V y condiciones galvanostáticas de 30 mA, 40 mA, 50 mA, 70 mA, 80 mA y 90 mA durante 10 minutos, utilizando como catalizador KOH y cuatro electrodos de acero previamente activados con una separación de 3 mm entre ellos (superficie 73 cm2), se permitió obtener los parámetros termodinámicos como son la entalpia de la reacción, la Energía Libre de Gibbs, y la Entropía; además se estableció el potencial de trabajo experimental óptimo y la eficiencia energética cuando se utilizaba una membrana Nafion 117 activada (superficie 36 Cm2) y en ausencia de la misma.**

Palabras Clave **- Propiedades Termodinámicas, celda combustible (reactor electroquímico), Nafion 117, potenciostáticas, galvanostáticas.**

I. NOMENCLATURE

E0: Equilibrium potential (V).
W: Electrical work (J)
ΔS: Change in entropy (J / mol* K)
ΔH: Change in enthalpy (J/ mol)
ΔG: Change in Gibbs free energy (J/mol)

II. INTRODUCTION

HYdrogen cells is a technology that is gaining great importance because it allows us to get electricity with zero greenhouse gas emissions the planet greenhouse[1, 2]; hydrogen for the study will be obtained at laboratory scale, using a prototype fuel cell having as electrolyte and proton exchange membrane (Nafion 117) [3]. The thermodynamic parameters, such as enthalpy, Gibbs free energy and entropy are paramount analyzed in order to establish how efficient the

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cell hydrogen production, where the working potential plays an important role in determining of these parameters. The proton exchange membranes are widely used in such technologies as they present very special characteristics: such as its high mechanical strength and high glass transition temperature [4, 5]. These features make it resistant to a chemical attack (bases and strong acids) and allows the membrane to withstand high temperatures. Their high permeability to H+ ions but not electrons, they make an electrolyte be widely used in fuel cells [6].

Driving increases at low temperatures, this is due to the inability of the polymer to retain the liquid at elevated temperatures (above 100 ° C), it is the reason why the Nafion is used in hydrogen fuel cells with low operating temperatures [7, 8]. The Nafion 117 membrane to be a poliperflurocarbonate polymer with sulfonic groups, allowing you to have hydrophobic and hydrophilic characteristics [6, 9].

The hydrophobic property (the carbon-fluorinated chain) helps remove water produced as a result of oxidation of the fuel (hydrogen), so the excess moisture from the membrane [10] is avoided; hydrophilic property (SO3- of sulfonic acid groups) are responsible for the transport of ions (H+) through the membrane, in other words it is the proton charge exchange [6, 11].

The ideal performance of a fuel cell is defined by the Nernst equation [12]. Which provides the relationship between the ideal standard potential (E0) for the reaction of the cell and the ideal equilibrium potential (E) and came given by:

$$E = E^{o} + \frac{\Delta S}{2F}(T - Tref) + \frac{RT}{2F} \qquad (1)$$

Where:
E = Nernst potential (V).
E0: equilibrium potential (V).
F: Faraday constant = 96485.5 C / mol
ΔS: change in entropy (J / mol K)
(PH_2); (P O2) gas pressure of hydrogen and oxygen respectively (atm).
R: 8.31452 J / mol * K.
T: Operation Temperature Fuel Cell (K).
T_{ref}: Reference temperature (K).

The potential between the electrodes defined as E0 is defined as the quantitative measure of the maximum cell potential open [13, 14] circuit, and is given by the following equation:

$$E = \frac{\Delta G}{2F} \qquad (2)$$

Where:
ΔG: Change in Gibbs free energy of formation.
F: Faraday constant

This study aimed to determine the thermodynamic parameters and design a fuel cell pilot plant that will be used to generate hydrogen, electrolytically operating in potentiostatic and galvanostatic mode with a proton exchange membrane Nafion 117.

III. Product development

A. Experimental Procedure

a. Materials and methods:

Several prototype hydrogen production cell were constructed with different geometry, that way the cylindrical shape is the most efficient because the reactor no dead zones and oxidation-reduction reactions that take there are more homogeneous [15]. The Cell capacity was 1000 cm3 (1L) for deionized water; 0.2 g of KOH as a catalyst, and a proton exchange membrane (Nafion 117) with an surface of 36 cm2 [16, 17]. For the thermodynamic parameters of the hydrogen fuel cell (PEM), preactivated steel electrodes for a period of 72 hours were used, in 1M H2SO4 solution, to form an oxide film on the electrode surface, enabling better electrical conductivity and also protect the corrosion electrode [18, 19]. The Steel anode consists of two steel plates, each with an surface of approximately 73 cm2 al as the cathode but the latter with the difference, that is coated with the Nafion 117 membrane; besides the spacing between the electrodes is figure 1, 3 mm. In steel electrodes used in the prototype of the hydrogen fuel cell is observed [6].

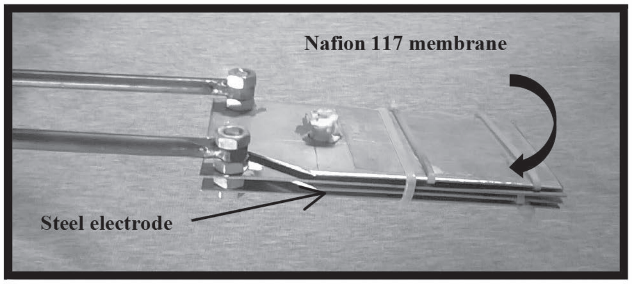

Fig. 1. Steel electrode with Nafion 117 membrane. Source: Author.

Figure 2 shows the Nafion 117 membrane used in the hydrogen production cell, which was activated by passing four bathrooms, each for an hour at a constant temperature between 70-80 ° C, in order to purify and hydrate (removing surface contaminants) and enrich it with ions [6, 20, 21]. Baths used were: 100 ml of hydrogen peroxide 3% by volume, 100 ml of deionized water, 100 ml of 2 M sulfuric acid and 100 ml with deionized water.

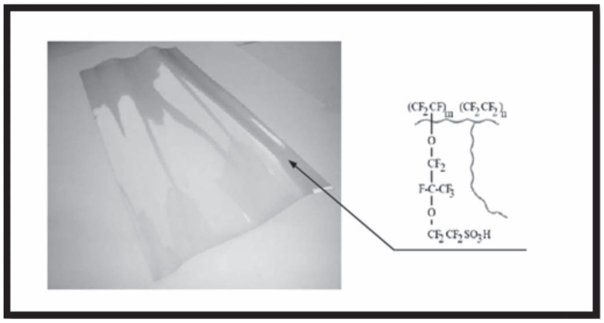

Fig. 2. Nafion 117 membrane. DuPont [6].

In the hydrogen production cell is carried out an electrolysis process which is summarized in the following chemical equation [11].

$$2H_2O_{(L)} \longrightarrow 2H_{2(g)} + O_{2(g)} \qquad (3)$$

The electrodes are connected to INTERFACE 1000 Gamry potentiostat and tests at constant potential (potentiostatic mode) 1 V, 1.23 V and 5 V and constant current (galvanostatic) 30 mA, 40 mA, 50 mA, 70 mA, 80 mA and 90 mA took for a period of 10min, and continuously sampled to determine the concentration and volume variation of the water within the cell. Figure 3 shows the general outline of the cell with all accessories [22].

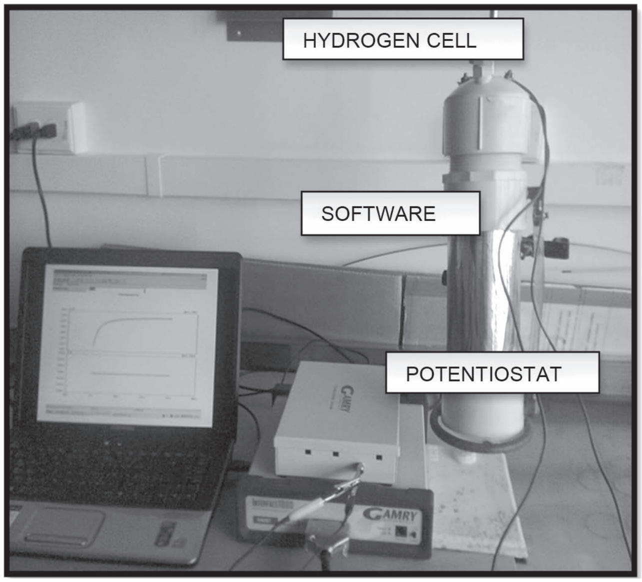

Fig. 3. General scheme of the fuel cell. Source: author.

IV. Results and ports discussions

a. Potentiostatic conditions

Figure 4 shows the behavior of the current density with respect to time to working potential of 1 V, 1.23 V and 5 V without Nafion 117 membrane, it can be inferred that the current density increases as the potential increases, presenting a reverse behavior with respect to energy efficiency [6, 23].

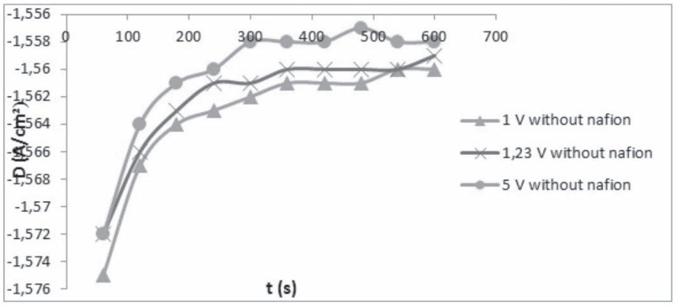

Fig. 4. Current density vs time potentiostatic conditions without Nafion 117 membrane.

For these devices wherein a hydrogen production process is performed, by electrolysis it is recommended the low current densities [13, 24], by the electrochemical parameters, we can interpret that the hydrogen production cell electrodes when steel is used, without Nafion 117 membrane potential is the recommended work 1V. The theoretical potential for hydrogen production cell is 1.23 V when platinum electrodes are used, where the experience takes this value tends to decrease [14], as was found with the experience gained. It is evident that there is a direct relationship between the potential applied to the cell and the current density, which indicates that as the current density increases, so does the potential, not to mention, it is advisable to afione a little work potential that promotes energy efficiency of the cell [6, 25]. So, the optimum working potential obtained experimentally Nafion 117 membrane when used, is 1V.

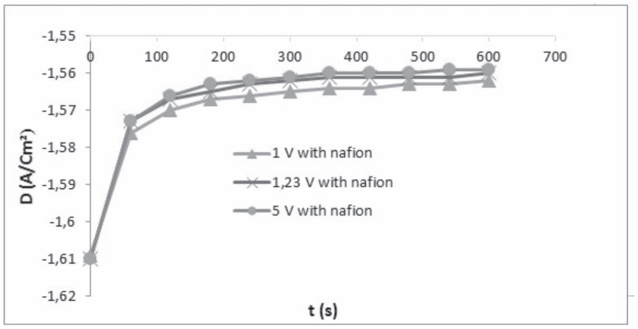

Fig. 5. Curves current density vs time to potentiostatic conditions Nafion 117 membrane.

Figure 6 shows the behavior of the cell without potentiostatic conditions with Nafion membrane 117 with the potential of 1 V. The proton conductivity of the membrane matches the chemical pretreatment prepared by the process performed hydration activation and sulfonic groups [7, 8]. Last was possible to be found, with decreasing current density, using a constant potential of 1 V.

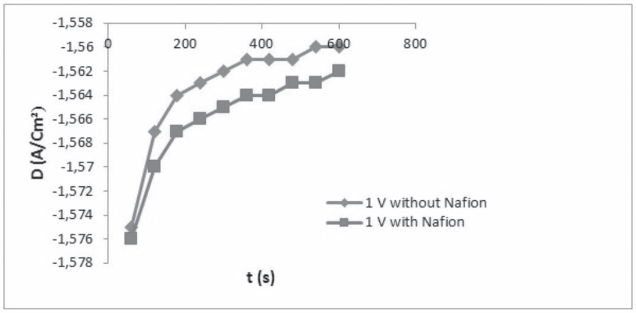

Fig. 6. Potentiostatic current density condition 1V.

Another important factor in decreasing the current density is the operating temperature of the cell (300 K); which is below the glass transition temperature (Tg) (270 ° C - 360 ° C for sulfonic groups and the polymer backbone 400-500) [7, 8, 26], allowing the membrane remains hydrated and does not lose its structural and functional properties.

b. Galvanostatic conditions

In Figures 7 and 8 constant behaviors shown intensities with and without membrane Nafion 117.

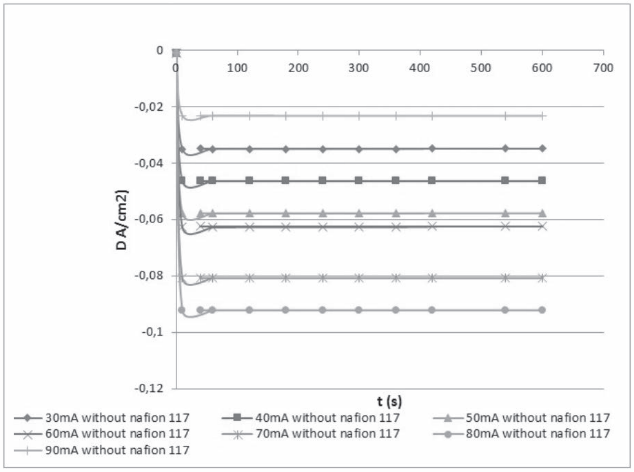

Fig. 7. Potentiostatic conditions without Nafion 117.

The values indicate that the current density decreases with increasing intensity of applied current, observing that in the first seconds of the reaction current density decreases and then remains constant throughout the reaction and no difference is found when the Nafion 117 membrane is used, or lack her [7, 8]. Finally it is concluded that galvanostatic conditions the reaction kinetics is controlled, so recommended by the electrochemical engineering is to characterize these devices to potentiostatic conditions [20, 21].

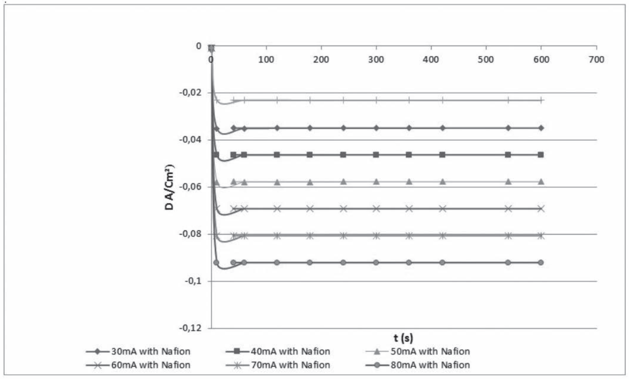

Fig. 8. Galvanostatic conditions with Nafion 117.

c. Energy efficiency potentiostatic conditions

The efficiency of a fuel cell is often expressed in terms of actual (experimental) and ideal (theoretical) voltage. Actual experimental voltage under ideal voltage (theoretical) due to losses associated with polarization and ohmic resistance of the cell. Therefore, the efficiency of a hydrogen fuel cell, can be expressed in function of the actual voltage (experimental) of the cell [2, 21, 27].

$$n = \frac{E\ exp}{Eteor} n_{teor} \tag{4}$$

Where:

E_{teor}: The theoretical ideal or theoretical potential of the cell that is equivalent to 1,229 V.

E_{exp}: The actual or potential is the experimental curves obtained characterization made potentiostatic conditions.

$n_{teór}$: = theoretical efficiency.

The obtained energy efficiency experimentally was 67%, indicating that the Nafion 117 membrane used in the production of hydrogen is sufficiently hydrated it is also an excellent electrolyte in conducting protons and the separation of hydrogen in the fuel cell, shown with the amount of hydrogen generated in the process [8, 21].

d. Efficiency using the calorific value of hydrogen

The calorific value of a fuel is defined by the number of units of heat released per unit mass or volume of fuel burned in a calorimeter agreed conditions.

Gross calorific (P.C.S) or HHV: Is the total heat of a fuel including latent heat of the water formed by combustion of hydrogen in said fuel.

Lower heating value (P.C.I.) or LHV: Efficiency in a hydrogen fuel cell can be calculated from the upper and lower specific heat of hydrogen, which is proportional to the current density [8, 14, 17].

If the gross calorific value used will be the following expression:

$$n = \frac{D}{1{,}482} \tag{5}$$

Where n is the efficiency of the hydrogen, D the current density.

But if the heating value of hydrogen is used the following expression [21, 27] is obtained:

$$n = \frac{D}{1{,}254} \tag{6}$$

In the process carried to fuel cell hydrogen potentiostatic conditions 1V, a curve of current density was obtained and used once the above expressions energy efficiency, shown in Figure 9 was determined. It is observed that the efficiency is higher when the hydrogen calorific value (LHV) is used; demonstrating again that the hydrogen fuel cell is more efficient at lower current densities.

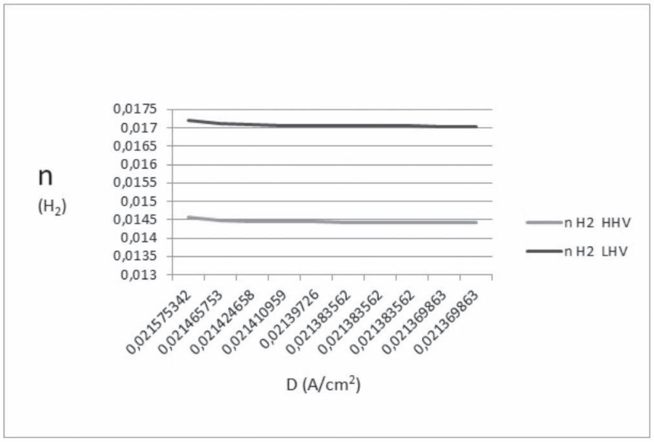

Fig. 9. Efficiency from the heating value of hydrogen.

e. Thermodynamics in cell hydrogen production

The thermodynamic variables allow quantify energy exchanges in the processes carried on the hydrogen cell. Development experience, there is no mixing between the liquid phase and the gas phase and thus a mathematical development was carried using tables where the values of enthalpy, entropy of water, oxygen and hydrogen is shown as inputs to calculate enthalpy (AHR) and the reaction entropy (ΔSR) [15, 19, 22]. Development experience, there is no mixing between the liquid phase and the gas phase and thus a mathematical development was carried using tables where the values of enthalpy, entropy of water, oxygen and hydrogen is shown as inputs to calculate enthalpy (AHR) and the reaction entropy (ΔSR).

$$H_2O_{(l)} \longleftrightarrow H_{2(g)} + \frac{1}{2} O_{2(g)} \qquad (7)$$

The values of enthalpy and entropy at standard conditions are in thermodynamic tables; 25 ° C and 1 atmosphere pressure [15, 19, 27].

TABLE I

ENTHALPY AND ENTROPY AT STANDARD CONDITIONS; 25 ° C AND 1 ATMOSPHERE PRESSURE

Compound	ΔH (J/mol)	ΔS (J/mol*ºC)
H_2O	-285000	69,9
H_2	0	130,6
O_2	0	205

SOURCE: MODIFIED BY AUTHOR

Enthalpy is the amount of heat energy of a substance. The enthalpy increase in a hydrogen production cell is given by the following expression [19, 22].

$$\Delta H_R(\mathrm{T,P}) = \Delta H_{H2}(T,P) + \frac{1}{2}\Delta H_{O2}(T,P) - \Delta H_{H2O}(T,P) \qquad (8)$$

Water that is in liquid state the enthalpy is determined using the following expression:

$$\Delta H_{H2O}(\mathrm{T,P}) = \Delta H_{H2O}(T,P) - H_{H2O}(T^{\ominus},P^{\ominus}) - \Delta H_{H2O}(T^{\ominus},P^{\ominus}) \qquad (9)$$

For hydrogen and gaseous oxygen obtained using expressions (8) and (9) respectively.

$$\Delta H_{H2}(\mathrm{T,P}) = H_{H2}(T,P) - H_{H2}(T^{\ominus},P^{\ominus}) - \Delta H_{H2}(T^{\ominus},P^{\ominus}) \qquad (10)$$

$$\Delta H_{O2}(\mathrm{T,P}) = H_{O2}(T,P) - H_{O2}(T^{\ominus},P^{\ominus}) - \Delta H_{O2}(T^{\ominus},P^{\ominus}) \qquad (11)$$

Now for the calculation of the enthalpy of reaction at a temperature other than the reference the following equations were used [21, 27].

$$H(\mathrm{T}) = a_j T + \frac{4}{5} b_j T^{5/4} + \frac{2}{3} c_j T^{3/2} + \frac{4}{7} d_j T^{7/4} \qquad (12)$$

$$S(\mathrm{T,P}) = a_j \ln T + 4 b_j T^{1/4} + 2 c_j T^{1/2} + \frac{4}{3} d_j T^{3/4} - R \ln T \qquad (13)$$

TABLE II

VALUES OF THE COEFFICIENTS OF THE EQUATIONS 12 AND 13

Compound	a_j	b_j	c_j	d_j
H_2O	180	-85,4	15,6	-0,85
H_2	79,5	-6,3	4,23	-0.19
O_2	10,3	5,4	-0,18	0

Taken from [21, 27] and modified by the author.

f. Calculation of increase in Gibbs Free Energy of reaction

Gibbs free energy (ΔG) is defined as that part of the total energy of a system can become work under isothermal conditions [19, 21].

Combining the entropy and enthalpy increase reaction increased Gibbs free energy of reaction is obtained [27].

$$\Delta G_R(\mathrm{T,P}) = \Delta H_{H2}(T,P) + \frac{1}{2}\Delta G_{O2}(T,P) - \Delta G_{H2O}(T,P) \qquad (14)$$

To the water in liquid state entropy is determined using the following expression:

$$\Delta G_{H2O}(\mathrm{T,P}) = \Delta H_{H2O}(T,P) + T(K)\Delta S_{H2O}(T,P) \qquad (15)$$

For hydrogen and gaseous oxygen obtained using expressions (14) and (15) respectively.

$$\Delta G_{H2}(\mathrm{T,P}) = \Delta H_{H2}(T,P) + T(K)\Delta S_{H2}(T,P) \qquad (16)$$

$$\Delta G_{O2}(\mathrm{T,P}) = \Delta H_{O2}(T,P) + T(K)\Delta S_{O2}(T,P) \qquad (17)$$

g. Calculation of the increase Entropy of the reaction.

Entropy is defined as a measure of thermodynamics, which represents the fraction of energy in a system that is not available to perform or perform a specific job. It is also defined as a measure of order or restrictions to perform a job that can be assimilated as a measure of molecular disorder of a system [19, 27].

Increasing the reaction entropy is determined using the following expressions [19]:

$$\Delta S_R(\mathrm{T,P}) = \Delta S_{H2}(T,P) + \frac{1}{2}\Delta S_{O2}(T,P) - \Delta S_{H2O}(T,P) \tag{18}$$

For hydrogen and oxygen generated gaseous expressions handled (17) and (18) respectively.

$$\Delta S_{H2}(\mathrm{T,P}) = S_{H2}(T,P) - S_{H2}(T^{\theta},P^{\theta}) - \Delta S_{H2}(T^{\theta},P^{\theta}) \tag{19}$$

$$\Delta S_{O2}(\mathrm{T,P}) = S_{O2}(T,P) - S_{O2}(T^{\theta},P^{\theta}) - \Delta S_{O2}(T^{\theta},P^{\theta}) \tag{20}$$

Determination of thermodynamic parameters:

The value of the free energy is experimentally found in the cell hydrogen production, taking into account the principle of thermodynamics which says work different minimum spanning work, it is necessary to provide for a process to occur not spontaneous at constant temperature and pressure, is equal to the increase in free energy of the process [13, 17, 19]. As hydrogen production cell is capable of exchanging one work (expansion work different) as electrical energy in this case is met:

$$\mathrm{W} = \Delta G_R \tag{21}$$

Where ΔGR is increased Gibbs free energy of reaction of water electrolysis. Furthermore, Faraday's law relates the electrical work and the conversion rate in terms of molar quantities. The expression of Faraday's law is [13, 22]:

$$Welec = qU = \mathrm{nFU} \tag{22}$$

TABLA III

EXPERIMENTAL DATA

Thermodynamic parameters	**Value**
Reaction Enthalpy (ΔH_R)	285,82 KJ/mol.
Gibbs free Energy (ΔG_R)	192,97 KJ/mol
Reaction Entropy (ΔG_R)	-3,43KJ/mol*ºC
Temperature	300 K

Experimentally an enthalpy of 285.82 kJ / mol was found, indicating that it is the energy available to perform a job. According to the thermodynamic theory, not all this energy is converted into work, but a portion is lost as Entropy and remains, it is useful to perform the work (ΔG) energy [17, 19, 27].

Increase the value of the enthalpy is a positive value, indicating that the process carried on the hydrogen production cell is endothermic; it means the reaction heat gains, favoring the production of hydrogen and reduction of water with zero emissions of greenhouse gases [17, 22, 27].

Experimental data show that 192.97 KJ / mol, it is useful for performing external work energy; that in accordance with the thermodynamic theory, electrical work, which results in electron flow [15, 27].

The positive increase in the value of free energy shows that the electrolysis reaction in carried fuel cell prototype can't be done spontaneously, it is necessary to apply activation energy for the reaction to occur [17, 19, 22].

V. CONCLUSIONS

The cylindrical shape of the cell for producing hydrogen, which was the best experimental result generated, since the electrolysis reaction is more homogeneous. The separation of the electrodes is 3mm in the minimum distance contributes to the decrease in current density and hence to lower the voltage applied to the cell.

A potentiostatic conditions with Nafion117 membrane was determined that the optimum working potential is 1 V, as we noted in the curves of current density vs. time, wherein a decrease in the density of the same is evidenced.

Parameters enthalpy, entropy and free energy shows that the hydrogen production cell process is endothermic, takes place also spontaneously and not more favorable the initial state than the final state of the system. Nafion 117 Membrane, besides proton conduction, it is also suitable to separate the hydrogen and oxygen in the fuel cell electrolyte, this is shown with a 67% efficient and the amount of hydrogen generated in the process.

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