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**HYDROGEN GENERATION BY ALUMINUM OXIDATION IN ALKALINE SOLUTION**

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**Abstract.** *The world economy's dependence on fossil fuels is continuously growing, primarily because of the increase of the economic development provided by those fuels. Given the frequent price oscillation and the environmental impact observed over the years, a discussion about ways to produce renewable and clean electrical and thermal energy has commenced to gain force. With the discovery of the fuel potential of hydrogen, a new challenge arises as scientists want to guarantee that its production takes place with the least possible impact on the environment. The most common ways to produce hydrogen gas are hydrocarbons reforming and electrolysis. In electrolysis, where the water molecule is broken into oxygen gas and hydrogen gas by the passage of an electrical current, the production cost is very high and demands a big amount of electrical energy that is usually obtained from fossil fuels. The hydrocarbon reforming is still the most used one and requires the exposure of the fuel to a high temperature water vapor, but despite its reduced production cost, it still releases carbon monoxide and dioxide, leading to the same environmental issue presented by fossil fuels. In this scenario, hydrogen production pathways that do not release or capture CO<sub>2</sub>, rely on renewable energy, and can deliver competitive hydrogen prices are needed. The hydrogen production from metal ways, as for example, the oxidation reaction of aluminum in an alkaline solution, can be an efficient and potentially clean way for hydrogen generation, as it is free of greenhouse gases emissions, it relies on an exothermic reaction, uses an easily recyclable metal (Al) as the raw material (reactant) and promotes the waste reduction. With all the above mentioned in mind, the objective of the herein study is to present a mathematical model to be used to develop a sustainable hydrogen reactor. The conceived hydrogen reactor will generate a series of experiments, where parameters will be varied to investigate how such variations (concentration of the alkaline solution and mass of aluminum) will interfere in the outcome of the reaction. A literature review will be the foundation to provide comparative data. The mathematical model will compute the energetic efficiency of the process and explore the potential of hydrogen as a fuel, used in electrical vehicles.*

**Keywords:** *Hydrogen generation, renewable energy, mathematical modeling, fuel cells.*

## 1. INTRODUCTION

The industrial revolution introduced to the market a more efficient and faster way of production, using steam engines, and later, oil machines. However, the economic growth became dependent on fossil fuels, creating political conflicts and territorial disputes (DAS e VEZIROGLU, 2001). The first oil crisis that happened in the late 70's reinforced the idea that fossil fuels are not a renewable source of energy, therefore, they are finite. That new mindset promoted a big fuel price oscillation which directly affected the world economy. The crisis created a discussion about different ways to generate energy that is sustainable and durable, mostly because of the consequences that the greenhouse gases release would cause to people and to the environment (OLIVEIRA, 2020). There was, then, an increase in the use of new energy sources, like hydraulic, wind, nuclear, biomass, geothermal energy, and others, that could, somehow, mitigate the predominance of only one type of fuel.

Hydrogen got more interest and space in this scenario seen as a revolutionary energy solution, having a 100% free of greenhouse gases emissions combustion (WANG, WAN e WANG, 2009). The hydrogen gas has a higher calorific value, compared with the common hydrocarbons (DOS SANTOS, 2003), in other words, a smaller amount (mass) of fuel can be used to obtain the same amount of energy, compared with other fuels. The use of hydrogen as a fuel also brings some obstacles related primarily to its commercial use. As an element with very low emissivity, its flame can not be seen in the daylight, emitting a weak light. This can be something positive when considering that the radiation transmitted is very low, but, on the other hand, turns this fuel way more dangerous in case of accidents, since the flame can not be seen easily (ESTEVAO, 2008). Another very important aspect is that hydrogen has a very small ignition energy compared with other fuels.

The applicability of the hydrogen as a fuel also must take into consideration its production. Since hydrogen is very reactive, it is not found in its molecular form, being always linked with other elements. This means that its production demands a certain amount of energy. Those processes can also release greenhouse gases. One of the most known methods of production is the water electrolysis, which is defined by the breakdown of water molecules, releasing gas hydrogen and oxygen (KNOB, 2013). Hydrogen can also be produced by hydrocarbon reforming, which consists in exposing the fuel in high temperatures, breaking down the molecule into different sub products, hydrogen being one of them. This technology, despite being the most used one, is still very aggressive to the environment due to carbon monoxide emission.

In this scenario, the hydrogen generation by the oxidation of aluminum in alkaline solution, got attention from the scientific community, mainly because of its potential to be a route of efficient and sustainable hydrogen generation without emitting greenhouse gases. It does not need an energy input to take place, the reaction between water and aluminum is spontaneous and also has an easily recyclable metal as the raw material (reactant), contributing to the reduction of the waste discharged in nature. The objective of this study is to develop a sustainable hydrogen reactor, and by its mathematical model to calculate the energetic efficiency of the process, and apply this analysis to generation and use of hydrogen as a fuel for electric vehicles.

## 2. FUEL CELLS AND ANALYSIS OF THE HYDROGEN GENERATION

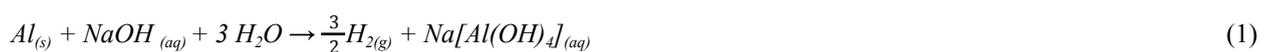
Fuel cells are devices that convert chemical energy into electricity by electrochemical reactions producing water and heat. In this study, we present the mathematical model of the electrochemical reaction of an Alkaline Membrane Fuel Cell (AMFC), where the electrolyte is an alkaline solution of potassium hydroxide. Chromatography paper is soaked in the alkaline solution (electrolyte) and is let to drip so the excess of the solution can be removed. The solution in the porous of the chromatography paper will allow the ions  $OH^-$  to move freely from the anode to the cathode. The Eq. (2) and (3) show the reactions occurring at the anode and cathode of the AMFC, respectively.

Since hydrogen is the fuel used in low temperatures fuel cells (AMFCs and PEMFC - Polymer Electrolyte Membrane Fuel Cells), we also conducted a study about hydrogen generation through the chemical reaction between water and aluminum. Fuel cells already play a major role in power generation using renewable fuels, however more has to be researched about methods of generating hydrogen in a sustainable way. With that in mind, we herein present a study about the aluminum alkaline oxidation for hydrogen generation, a brief literature review about the chemical reactions involved and a mathematical model of a fuel cell.

### 2.1 Hydrogen generation by metal ways

Hydrogen generation by the aluminum oxidation in an aqueous solution has been a promising alternative to hydrogen generation. Aluminum is the most abundant metal on earth and due to its chemical and mechanical properties, it is often used in the fabrication of packages and household utensils. Since the recycle process of this metal is one of the most known and applied worldwide, the aluminum used in the hydrogen generation reaction can be provided by its recycled form, making it easier to get the raw material necessary and at the same time helping the environment and making it to be considered sustainable (CASSANELLI, 2016). One of the most favorable economic aspects for the application of this method is the facility to obtain the reagents, in this case water and aluminum, using sodium hydroxide as catalyst, that can be considered low cost when compared to the raw materials for other types of hydrogen production. Besides that, aluminum hydroxide has a considerable industrial relevance and the system does not need expensive equipment to work, which decreases even more the cost of the whole process (HIRAKI, 2007; AKIYAMA, 2009).

The hydrogen generation consists in reacting water and aluminum, oxidizing the metal and producing hydrogen. It is an exothermic reaction releasing 853 kJ (CASSANELLI, 2016). The aluminum in contact to the atmospheric air oxidizes producing aluminum oxide, a thin layer that isolates the aluminum from the outside, requiring a catalyst that will minimize the reaction time that naturally would initiate after a few hours. The catalyst increases the efficiency of the reaction. The catalyst used in this study is NaOH. Sodium hydroxide is largely used in industry making its acquisition easier, but other basic catalyzes and chemical hydrides can also be used. NaOH dissociates when in aqueous solution, forming  $OH^-$  ions that block aluminum oxide formation, exposing the metal in contact to the water and initiating the reaction. Therefore, the global reaction is characterized by (HSIEH; HER; CHEN, 2012):



The Eq. (1) is a spontaneous reaction, which means that it does not need an energy input to happen. It also produces hydrogen with a high degree of purity, around 99%, (MARTÍNEZA et al., 2005), excellent to be used into fuel cells. The reaction is directly affected by the contact area between the metal and the alkaline solution, and, because of this, it is important to keep the aluminum sliced in smaller pieces to obtain a better outcome. Sodium aluminate is also a product of the reaction and has various applications in industry, and because of that, it can be recycled. It is used in the fabrication of firebricks and it is added as an additive to cement used in cold weather, making the drying process easier, it also has applications in pharmaceutical and textile industry (WANG et al., 2009). Furthermore, the aluminate can be dissociated to obtain again sodium hydroxide, reutilized in future reaction, or it can give rise to aluminum hydroxide, used in makeup, skin care products and all kinds of cosmetic products production. It even can be used as treatment for some diseases and to prepare vaccines, like A and B hepatitis and tetanus (PEREIRA, 2017). The dissociation reaction for sodium aluminate is:



### 2.2 Chemical reactions

To analyze the influence of the NaOH concentration in the hydrogen gas production, three different sodium hydroxide concentrations were tested, 2 mol/L, 3 mol/L and 4 mol/L, assessed for 5g, 8g and 10g of aluminum. To prepare the solutions of 2, 3 and 4 mol/L, it was necessary to weigh in the precision balance 20g, 30g and 40g of sodium hydroxide, respectively, for 250 ml of distilled water. After that, each sodium hydroxide grammage is separated in a beaker with the correct identification and mixed with a small part of the distilled water so that the hydroxide can be

dissolved. Because the reaction is exothermic, it was necessary to wait a few seconds to pass the solution for the volumetric flask, where the rest of the water was mixed. With the solutions ready, it can be reacted with the same amount of aluminum to observe the reaction behavior for each concentration. On the other side, to observe the influence of the aluminum in the reaction and to analyze how the reaction behaves during the metal oxidation, the reaction was made for each NaOH concentration, 2, 3 and 4 mol/L, varying the aluminum amount in 5g, 8g and 10g. Figure 1 shows the shape of the aluminum used as the reactant of the Eq. (1) and the reactor generating hydrogen.



(a)



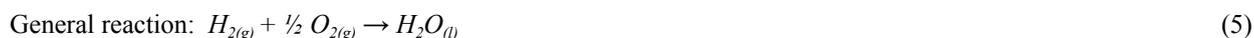
(b)

Figure 1. (a) Sample of the aluminum used in the hydrogen generation reaction and (b) reactor generating hydrogen.

### 2.3 Alkaline Membrane Fuel Cell Mathematical Model

The mathematical modeling presented herein was previously introduced by Furlan et al. (2020) and we will present below its main concepts. The software chosen to perform the modeling was Interactive Thermodynamics, mainly because its simple and convenient interface and also because the simplicity to conduct parametric studies.

To be able to analyze the thermal behavior of the hydrogen production reaction, it is necessary also to know and dominate the oxidoreduction that happens inside the fuel cell, in which the hydrogen is used. The electrochemical reactions taking place in the AMFC are the following:



This way, to be able to predict the thermodynamic behavior of the aluminum oxidation reaction that produces hydrogen, it is necessary to do the thermodynamic modeling of the general reaction and the anode reaction, investigating the energetic efficiency of an alkaline fuel cell, working under different temperatures.

The equations needed to describe the thermodynamic behavior of each component present in the reaction are the following, with R being the universal gases Constant and  $\Delta n_g$  being the electrons variation in gaseous state:

Enthalpy, described for:

$$\Delta H_{(total)} = \sum \Delta H_{(reag.)} - \sum \Delta H_{(prod.)} \quad (6)$$

Entropy, described for:

$$\Delta S_{(total)} = \sum \Delta S_{(reag.)} - \sum \Delta S_{(prod.)} \quad (7)$$

Enthalpy variation, described for:

$$\Delta H = \Delta U + \Delta n_g RT, \quad (8)$$

Free Gibbs energy, described for:

$$\Delta G_{(total)} = \sum \Delta G_{(reag.)} - \sum \Delta G_{(prod.)} \quad (9)$$

Free Gibbs energy variation, described for:

$$\Delta G = \Delta H - T\Delta S \quad (10)$$

The value for each one of these properties can be seen in Table 1, above, modified from (CHEN, et al, 1991; LIDE, 2004):

Table 1. List of thermodynamics properties in standard state: (Furlan *et al*, 2020)

Compound name	Molecular formula	$\Delta H^0$ (kJ/mol)	$\Delta G^0$ (kJ/mol)	$S^0$ (J/mol · K)	$C_p$ (J/mol · K)
Aluminum	Al <sub>(s)</sub>	0	0	28.30	24.4
Aluminum Hydroxide	Al(OH) <sub>3(s)</sub>	-1284	-1306	71	93.1
Sodium aluminate	Na[Al(OH) <sub>4</sub> ] <sub>(aq)</sub>	-1742.84	-1567.18	161.35	-
Sodium hydroxide	NaOH <sub>(aq)</sub>	-469.15	-419.20	48.1	-102.1
Oxygen gas	O <sub>2(g)</sub>	0	0	205.14	29.33
Hydrogen gas	H <sub>2(g)</sub>	0	0	130.680	28.84
Hydroxyl	OH <sub>(aq)</sub>	-229.99	-157.24	-10.75	-148.5
Water	H <sub>2</sub> O <sub>(l)</sub>	-285.83	-237.14	69.95	75.35

With that, the calculation of the reaction efficiency of Eq. (5), which for being a chemical reaction that happens inside a fuel cell, is calculated as:

$$\eta = \frac{\Delta G}{\Delta H} \quad (11)$$

And for the reversible potential created by the fuel cell, it is:

$$E_r = - \frac{\Delta G}{n_e F} \quad (12)$$

Equations (6)-(12), and Table 1 are implemented into IT according to the flowchart of Figure 3.

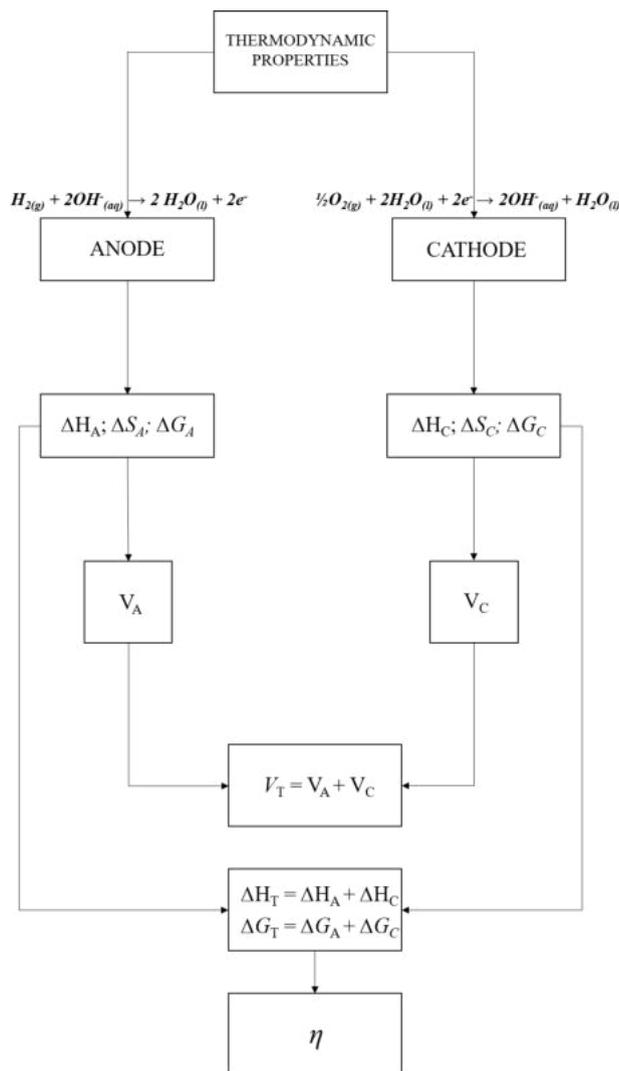


Figure 3. Flowchart of the process implemented in IT.  $V_A$  and  $V_C$  are the reversible potential for anode and cathode respectively,  $V_T$  is the total reversible potential. The subscripted A and C represent the anode and cathode, respectively. (Furlan *et al.*, 2020)

The code was executed following the steps described in Fig. (3), in order to obtain thermodynamic properties and the efficiency of a fuel cell system. Table 2 presents the results obtained for temperatures in the range 298.15K to 373.15K, using Eq. (3-5):

Table 2. Enthalpy variation ( $\Delta H$ ), Free Gibbs energy variation ( $\Delta G$ ), and maximum efficiency ( $\eta$ ) vs temperature (K) according to Eq. (3) and Eq. (4): (Furlan *et al.*, 2020)

T (K)	$\Delta H$ (kJ/mol)	$\Delta G$ (kJ/mol)	$\eta$ ( $\Delta G/\Delta H$ )
298.15	-285.8	-237.1	0.8296
323.15	-285.8	-237.9	0.8323
348.15	-285.8	-238.7	0.8350
373.15	-285.8	-239.4	0.8377

### 3. RESULTS

With the intention to analyze the reaction behavior with the aluminum amount variation used to the same NaOH concentrations, 2, 3 and 4 mol/L, the reaction has observed for 5g, 8g and 10g of aluminum, under the concentrations, 2, 3 and 4 mol/L of NaOH. The Fig. (4)-(6) below represent temperature vs time for each amount of aluminum used.

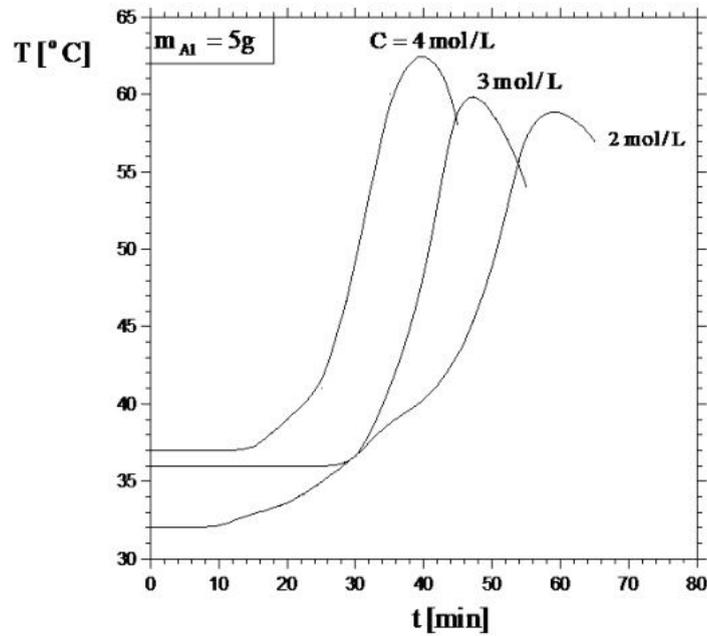


Figure 4. Experimental temperature variation for 5g of aluminum and varying NaOH concentrations.

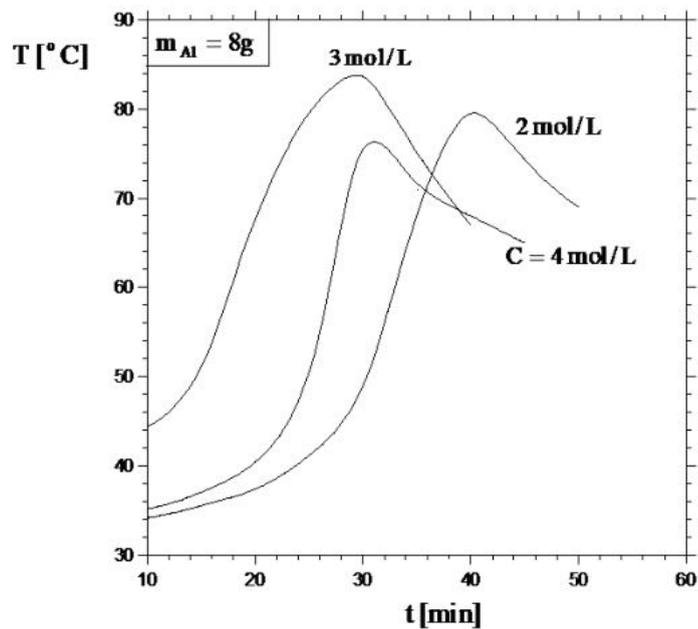


Figure 5. Experimental temperature variation for 8g of aluminum and varying NaOH concentrations.

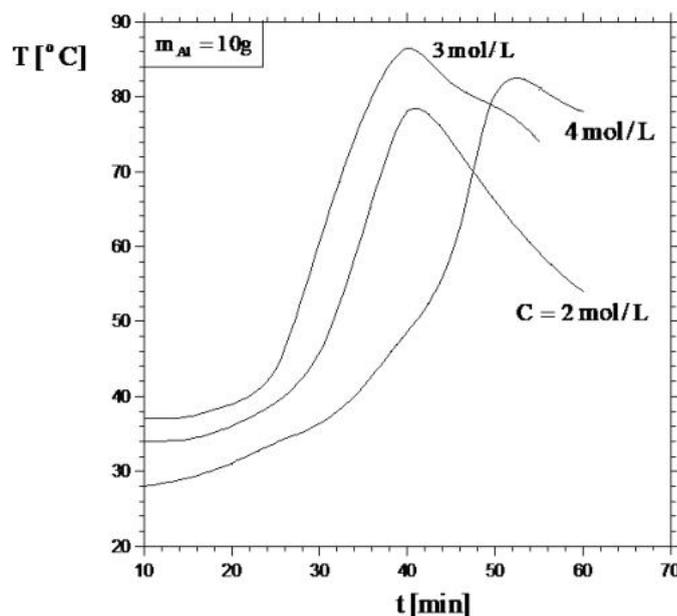


Figure 6. Experimental temperature variation for 10g of aluminum and varying NaOH concentrations.

According to Figures 4-6, it is possible to state that aluminum is the limiting reactant in the hydrogen generation reaction. It can be seen by the similarity of the curves of each graph when varying the alkaline solution concentration (C). For the same amount of aluminum, the time needed to obtain the highest temperature varies and hence, the maximum hydrogen production. Still, the time span is very short, varying two to ten minutes with the concentration increase, depending on the amount of aluminum. Another factor that can affirm the limiting character of aluminum is the fact that the highest temperatures reached during the reaction is related to the aluminum amount and not the concentration of the solution. It is possible to observe that for 5g of aluminum, the highest temperature reached is near 60°C, while for 8g is around 80°C and 10g, around 85°C.

It was possible to observe, on the other hand, by Fig. 4, 5 and 6, that a higher initial temperature increases the reaction velocity, making the aluminum consumption faster.

#### 4. CONCLUSIONS

Considering that the type of cell used in this study is a low temperature cell (alkaline membrane fuel cell), since high temperatures have a negative effect on the performance of the cell, the most indicated sodium hydroxide concentration is 2 mol/L. This can be explained because in this concentration, the maximum temperatures vary around 60 and 70°C, while for the other concentrations, this value exceeds 80°C. However, further studies need to be conducted to check what is the effect of the temperature of the hydrogen in the performance of the fuel cell. In this context, the control of the exothermic behavior of the reaction is essential to its optimization. That means that Eq. (1) is extremely exothermic, releasing a lot of heat, causing the system temperature to increase.

The use of hydrogen as a fuel still brings a lot of technical and commercial issues, the present study is intended to advance science and the technology towards a more affordable and sustainable way to generate hydrogen to be used in hydrogen and battery electric vehicles.

#### 5. ACKNOWLEDGMENTS

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