ORIGINAL PAPER

EXPERIMENTAL STUDY OF HYDROGEN PRODUCTION THROUGH ELECTROLYSIS OF ALKALINE SOLUTION USING OXIDES OF ZINC-IRON ALLOY AS CATHODE

FATIMA EZZAHRA CHAKIK[1,2]*, MOHAMMED KADDAMI[1], MOHAMMED MIKOU[2]

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Abstract. *Electrolysis of water is a promising option to produce hydrogen with high purity using solar energy. The performance of electrolysis of water is influenced by several parameters. The main objective of this work is to study the production of hydrogen through electrolysis of water on testing new cathode alloy: Zn-Fe. Using experimental design, a regression function relating the volume of hydrogen produced and three factors: input voltage, input amperage and electrolyte concentration was developed. The results revealed that all factors studied have a positive effect on volume of hydrogen produced. The interactions between these factors are not negligible and must be taken into account during the production of hydrogen. The maximum volume 14.2 mL of H_2 at 4 min is obtained when the three factors are at their high levels: 5 V, 0.5 A and 30 g/L of NaOH.*

Keywords: *Electrolysis of water, hydrogen, cathode alloy, experimental design.*

1. INTRODUCTION

Hydrogen production from water electrolysis, using solar energy, is a promising option to develop new energy systems based on renewable resources. In the past several decades, the increase in energy consumption has raised the exhaustion of the globe's reserves of natural resources. Today our economy and technologies mainly depend on natural resources, which are not replaceable, due to industrializations and population growth. Approximately 80% of our energy consumption comes from fossil fuels (Oil, Natural gas and Coal) [1]. The use of fossil fuels presents many problems; Fossil fuel reserves are limited stock (It estimated 1688 Billion barrels for Oil, 6558 Trillion Cubic Feet for Gas and 891 Billion tons for Coal) [2]. Moreover, the use of fossil fuels harms the environment. Indeed, the burning fossil fuels leads to air pollution and global warming. Therefore, renewable energy plays an important role in reducing the use of these fossil fuels, which are major sources of carbon dioxide emissions. Renewable energy has an intermittent nature; it should be combined with a storage system for an effective utilization. Consequently, one of the ideal ways proposed, is the transformation of solar energy into chemical energy stored in the form of hydrogen, through water electrolysis [3].

Electrolysis of water is the process of splitting water into hydrogen and oxygen gas using two electrodes connected with a power supply. This process has attracted attention of many researchers because of their simplicity, ecological cleanliness and it easy maintenance [4]. The hydrogen produced with this technology has a high purity 99.999 vol% [5].

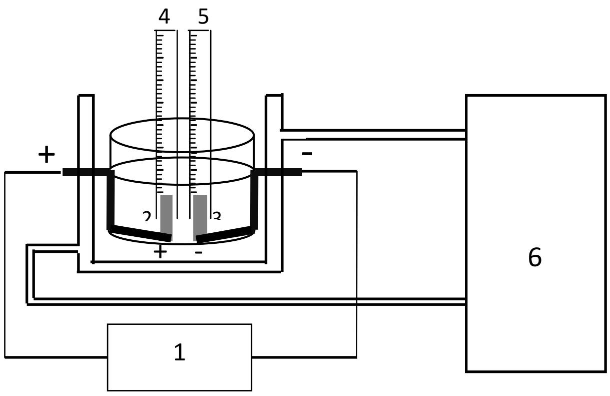
[1] University Hassan 1st, Faculty of Science and Technology, Laboratory of Physical Chemistry of Processes and Materials, BP 539 Settat, Morocco. E-mail: f.chakik@uhp.ac.ma.
[2] University Hassan 1st, Faculty of Science and Technology, Laboratory of Analysis of Systems and Information Processing, BP 539 Settat, Morocco.

The performance of electrolysis of water is affected by several factors such as electrodes materials, electrolyte temperature, electrolyte type and concentration, pressure, space between electrodes etc. For example, in the reference [6], the authors revealed that the optimum condition on hydrogen production, during water electrolysis using Ni-Cr-Fe alloy as electrodes, was found by decreasing the space between electrodes. Sellami et al. [7] suggests that better hydrogen production is achieved with the use of potassium hydroxide (KOH) as an electrolyte. In this context, the present works aims to study the production of hydrogen through electrolysis of water with new cathode alloy: Zn-Fe. The effect of some parameters in hydrogen production is evaluated by using experimental design.

2. MATERIALS AND METHODS

2.1. ELECTROLYSIS CELL

In the present study, we used the experimental electrolysis cell described in our previous paper [8] and illustrated in Fig. 1. The cell consists of a generator Aim-TTi CPX 420 W per output, two electrodes (anode and cathode), tubes to collect gas produced and thermostat to fix cell temperature. At the cathode, there is a production of hydrogen and at the anode; there is a production of oxygen gas. The distance between the two electrodes has been optimized at approximately 1.5 cm. Electrode's surface submerged into the electrolyte is fixed at 5 cm^2.



1 Power Supply
2 Anode
3 Cathode
4 Tube to collect oxygen gas
5 Tube to collect hydrogen gas
6 Thermostat

Figure 1. Experimental set-up for electrolysis of alkaline water.

2.2. ELECTRODE PREPARATION

For the anode, we have used a plate of Nickel. Due to its high activity and availability, Nickel is the most material employed as anode [9]. For the cathode, the metals used were Zinc (Zn 98%) and Iron (Fe 99%), supplied by Sigma-Aldrich. The cathode is prepared according to the procedure described in our previous workd [8]. A binary alloy of about 3 g and composed of 95% of Zn and 5% of Fe (mass %), was accurately weighed. The simple was homogenized in a mortar, and then compacted using a hydraulic press at 10 MPa for 1 min at room temperature. The obtained cathode was heat treated under standard atmosphere at 800 °C for 2 h.

2.3. DESIGN OF EXPERIMENTS

In order to evaluate factors that influence the volume of hydrogen produced, a full factorial experiment was designed. The full factorial design is a good technique to study the interactions between parameters influencing a response at a reduced number of experimental. This technique is also applied for optimization and modeling [10]. In the case of full factorial design, the number of experimental runs that has to be carried out for a two level and n factors: is 2^n runs [11]. In this study, three factors were chosen: input voltage, input amperage and electrolyte concentration. Each factor was studied at two levels: low level and high level.

To analyze the full factorial design, the experimental measurement (uncoded units) were transformed into coded units [12]. Therefore, the factor levels were coded as −1 (low) and 1 (high). The response was expressed as the volume of hydrogen produced at 4 min of time reaction. The cell temperature was fixed at 25 °C during experiments. Sodium hydroxide NaOH (Sigma-Aldrich 98%) is chosen to use as en electrolyte throughout the experiments. Table 1 shows the factors and levels used.

Table 1. Factors and levels used in full factorial design.

Parameters	Symbol	Low level (-1)	High level (+1)
Input voltage (Volts)	X_1	3	5
Input amperage (Amps)	X_2	0.2	0.5
Electrolyte concentration (g/L)	X_3	10	30

MINITAB 17 Software was used to create and analyze the experimental data. The polynomial model proposed to define the relation between the response Y (volume of hydrogen / 4 min) and the three factors: input voltage (X_1), input amperage (X_2) and electrolyte concentration (X_3), is described according to full factorial design [12] by Equation (1):

$$Y = a_0 + a_1X_1 + a_2X_2 + a_3X_3 + a_{12}X_1X_2 + a_{13}X_1X_3 + a_{23}X_2X_3 + a_{123}X_1X_2X_3 \quad (1)$$

where a_0 is the average response in a factorial experiment, a_i and a_{ij} are the regression coefficient.

3. RESULTS AND DISCUSSION

3.1. X-RAY DIFFRACTION ANALYSIS OF Zn-Fe CATHODE

The cathode (Zn95%Fe5%) prepared in this study and heat treated under standard atmosphere, is characterized by X-ray diffraction (D2 PHASER diffractometer (CuKα radiation (λ=1.5418 Å; 40KV and 30 mA)). A software HighScore Plus version 3.0e [13] is used to identify the obtained phases. Fig. 2 presents the XRD patterns obtained for (Zn95%Fe5%) cathode. The result shows the formation of five compositions: $ZnFe_2O_4$ (JCPDS 01-079-1150 [14]), ZnO (JCPDS 01-070-2551 [15]), FeO (JCPDS 00-046-1312 [16]), Zn (JCPDS 00-004-0831 [17]) and Fe (JCPDS 01-089-4186 [18, 19]).

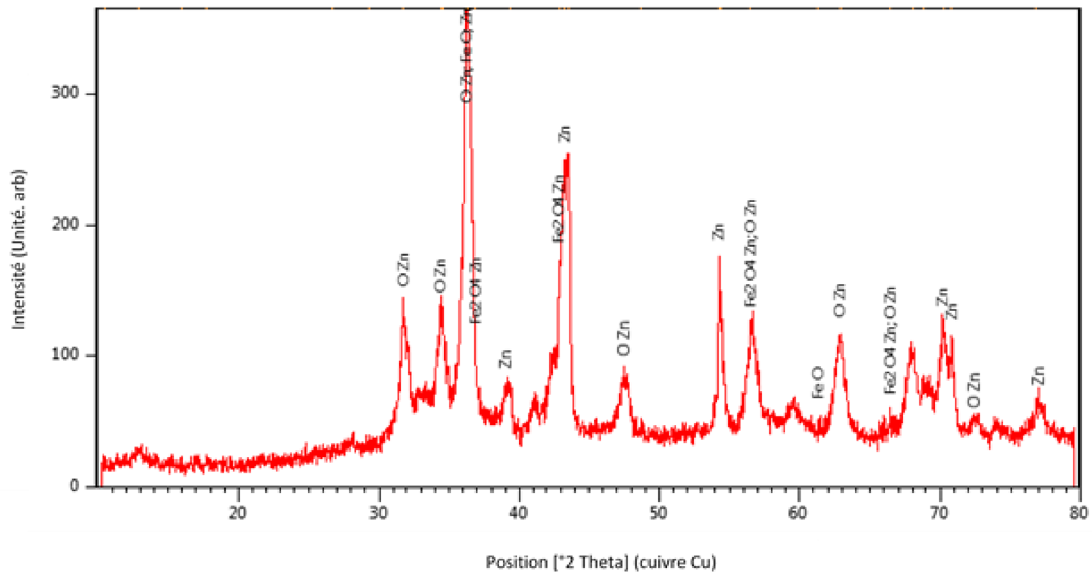


Figure 2. X-ray diffraction analysis of Zn95%Fe5% (mass %) cathode.

3.2. MODELIZATION OF VOLUME OF HYDROGEN PRODUCED

The design matrix of coded values for the factors (X_1, X_2 et X_3) and the response Y in terms of the volume of hydrogen produced at 4 minutes of time reaction for all experimental runs, are shown in Table 2. A linear regression model was fitted for the experimental data, using MINITAB 17. Equation (2) gives the coded model.

$$Y = 4.688 + 2.863X_1 + 1.712X_2 + 0.9375X_3 + 2.337X_1X_2 + 0.4125X_1X_3 + 0.5625X_2X_3 + 0.6875X_1X_2X_3 \quad (2)$$

Table 2. Experimental design matrix and results.

Run order	X_1	X_2	X_3	Y
1	-1	-1	-1	1.8
2	1	-1	-1	3.4
3	-1	1	-1	0.8
4	1	1	-1	9
5	-1	-1	1	3.1
6	1	-1	1	3.6
7	-1	1	1	1.6
8	1	1	1	14.2

To validate the regression model, other experimental data obtained under other reaction conditions and within the lower and upper limits of each factors, are used. Table 3 reports the obtained results.

Table 3. Tests to validate the regrassion model obtained.

Test	X_1	X_2	X_3	$Y_{measured}$	$Y_{predicted}$
1	0.00	-0.33	0.00	4.2	4.1
2	1.00	0.00	0.00	7.7	7.6
3	-1.00	0.33	1.00	2	2.1
4	0.00	-1.00	1.00	3.6	3.4

Fig. 3 shows a clear comparison of the measured volumes and those predicted by the regression model. The correlation coefficient obtained for the validation data is around

R_v^2 = 99.7%, which means a good adjustment of the regression function to the experimental data. From these experimental results, we can conclude that the regression model can be used to predict and measure the volume of hydrogen within the domain of variation of each factors studied.

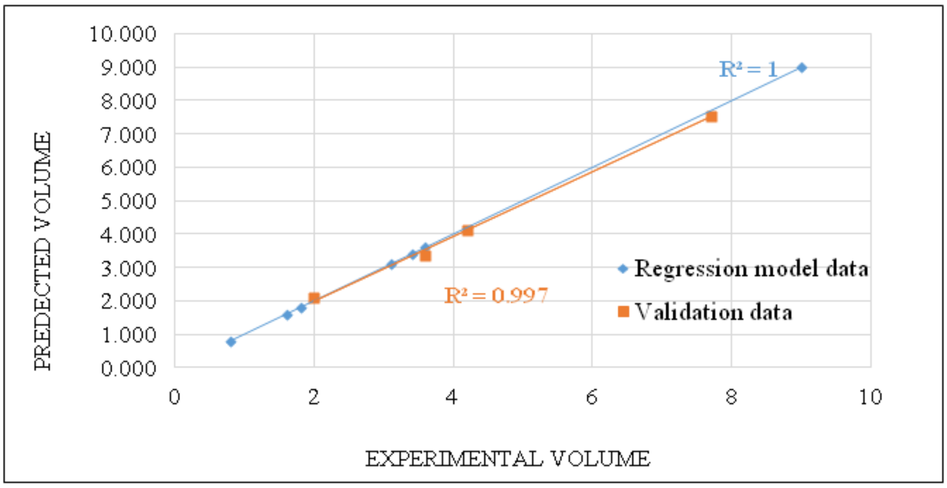


Figure 3. Comparison between observed and predicted volume in the case of Zn95%Fe5%.

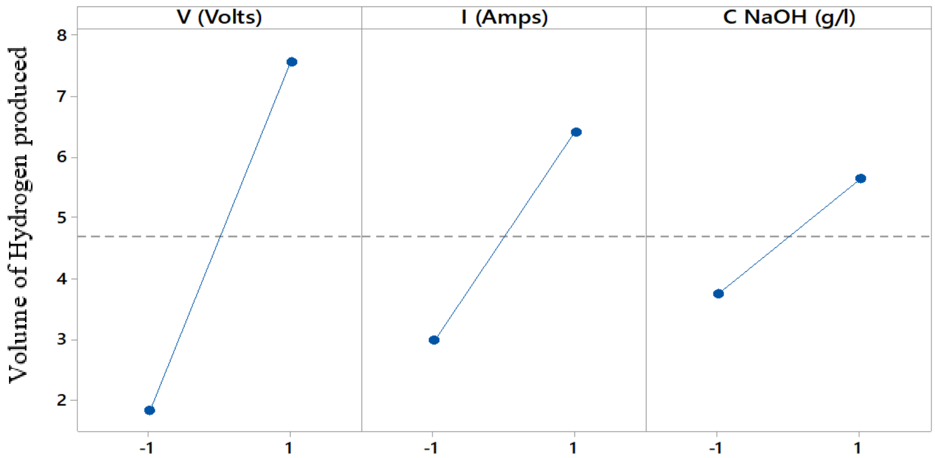


Figure 4. Diagram of the main effects of three factors (input voltage, input amperage and electrolyte concentration) on hydrogen production.

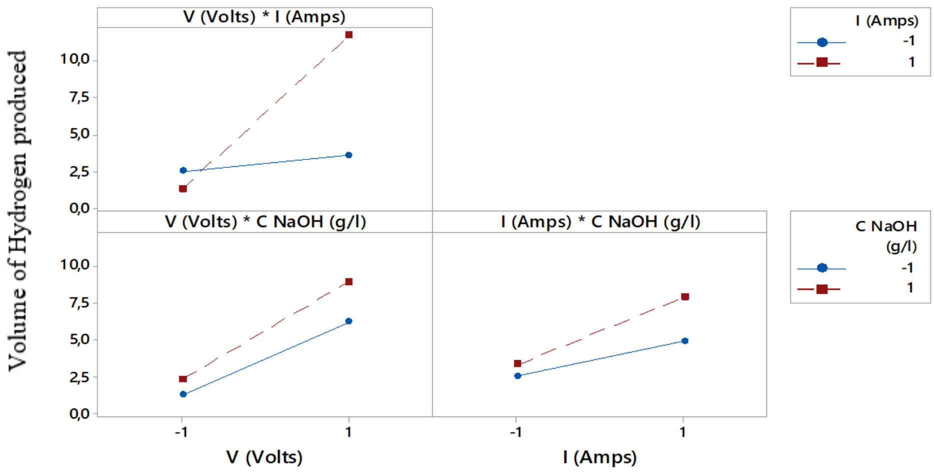


Figure 5. Diagram of interactions effects between three factors (Input voltage, input amperage and electrolyte concentration) on hydrogen production.

Figs. 4-5 illustrate the effects of each factor and their interactions respectively on the volume of hydrogen produced. The input voltage is the most important factor that influence the volume of hydrogen. The electrolyte concentration seems do not have a strong effet compared to the two other factors. However, to have a complete interpretation of the results, we have taken the interactions into account, and from Fig. 5, it can be seen that they are not negligible. The interaction between the voltage and amperage is very significant. A change in input amperage at higher level of voltage has a strong effet on volume of hydrogen than at lower level. In addition, the same remark was observed for voltage-concentration and amperage-concentration interactions. Therefore, the production of hydrogen is influenced by the interaction between all parameters studied.

4. CONCLUSIONS

In this study, the production of hydrogen by electrolysis of alkaline solution was studied using new cathode alloy. The results show that the cathode alloy Zn95%Fe5%, heat treated under standard atmosphere, is very active and produce more hydrogen at different reaction conditions. By applying the experimental design, a regression function that related the volume of hydrogen produced with three factors: voltage, amperage and electrolyte concentration was developed. The results revealed that the volume of hydrogen grows larger as the voltage and amperage increase. The interactions between factors studied are not negligible and must be taken into account during the production of hydrogen. The maximum volume is attained when the three factors are at their high levels: 5 V, 0.5 A and 30 g/L NaOH.

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