



Analysis of Electrodes Consumption via the Electrocoagulation Treatment of Lead Removal from Simulated Wastewater

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ABSTRACT

The aim of the present work is to analyze the phenomenon of electrodes consumption (i.e. anode and cathode electrodes) when it used in a batch electrocoagulation reactor for the removal of lead from a synthesis wastewater. The studied operational parameters were the electrolysis time (5-60) minute, pH value (2-12), applied current (0.2-2.6) Amperes, and the initial concentration of lead and the stirring speed were fixed at 155ppm and 150rpm respectively. Aluminum concentric tubes were used as electrodes by making the outer and inner as the anode electrode and the rest in between as the cathode electrode. Statistica-10 and Minitab-17 statistical programs were used to analyze responses via response surface methodology (RSM) technique to find the correlations that relate to all variables. The results show that both of electrodes had consumed and increased or decreased in that way belong to the operating variables. Applied current and pH value of the simulated solution were founded as the main effect of electrode consumption.

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تحليل استهلاك الأقطاب الكهربائية أثناء المعالجة بالتخثير الكهربائي لإزالة الرصاص من مياه الصرف المحاكاة

الخلاصة

ان الهدف من الدراسة الحالية هو لتحليل ظاهرة استهلاك الاقطاب الكهربائية (الأنود والكاثود) والتي تُستخدم في مفاعل التخثير الكهربائي ذو الدفعات عند ازالة الرصاص من مياه الصرف المصنعة. ان المتغيرات التشغيلية قيد الدراسة قد كانت زمن التلامس (٥-٦٠) دقيقة والدالة الحامضية (٢-١٢) والتيار المسلط على الخلية (٢,٦-٠,٢) امبير وتم اعتماد قيم ثابتة للتركيز الاولي للرصاص وكذلك سرعة الخلط والتي كانت على التوالي 155 ملغم/لتر و 150 دورة بالدقيقة. تم استخدام اقطاب انبوبة متحدة المركز ومادتها من الالمنيوم بحيث كان الانبوب الداخلي والانبوب الخارجي يشكلان سوية قطب الأنود والانبوب الوسطي الذي بينهما يمثل قطب الكاثود. تم استخدام برامج احصائية لتحليل الاستجابات من خلال تقنية (RSM) لايجاد علاقات رياضية تربط كافة المتغيرات. أظهرت النتائج ان هنالك استهلاك في كلي القطبين الأنود والكاثود ويزداد ذلك او ينقص تبعاً للمتغيرات التشغيلية. ان التيار المسلط والدالة الحامضية يعتبران المؤثران الرئيسان في عملية استهلاك الاقطاب الكهربائية.

الكلمات المفتاحية

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Introduction

Despite the using of various methods to treat water and wastewater, the electrocoagulation technique has received a wide consideration attention due to its effecting in dealing with numerous pollutants [1] because of its low infrastructure requirements, ease of use and potential for scalability [2 and 3].

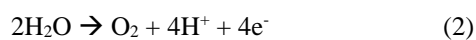
It is a clean electrochemical technique and could be classified as an environmentally friendly method [4] and so efficient method to recover heavy metals, that have a specific gravity greater than 5.0, from wastewater under appropriate restrictions [5 and 6].

Fundamentally, the main responsible effect of the electrocoagulation performance is the production of flocs $\text{Al}_2(\text{OH})_3$ as a result of the electrodeposition of aluminum ions from both of electrodes [7] as shown in Fig. 1.

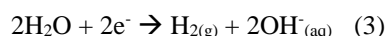
Moreover, flocs will work as an adsorbent in the solution to recover the contaminate from the wastewater [8] according to the following steps [9]:

- 1- Formation of flocs as a result of the oxidation and reduction operations on anode and cathode respectively as follow:

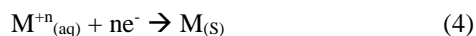
- At the anode electrode with metal M:



- At the cathode electrode:



- 2- Destabilization of the pollutants, particulate suspension, and breaking of emulsions. At the cathode surface:



- 3- Aggregation of the destabilized phases to form flocs.

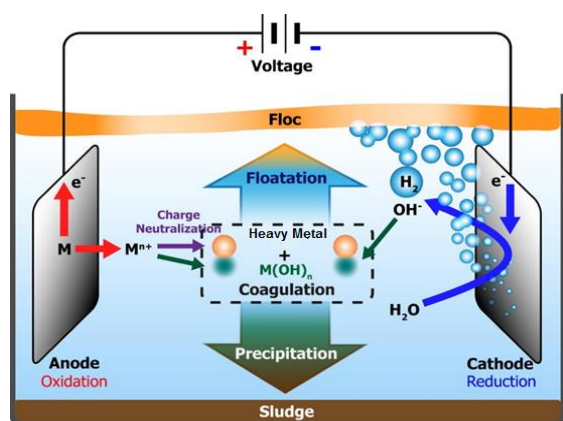


Figure 1: Schematic of electrocoagulation technique

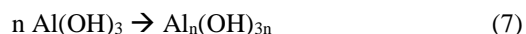
The electrodeposition of aluminum in the electrocoagulation cell is the main reason for the electrodes consumption along the duration of the experiments done [10].

Therefore, electrocoagulation method involves two essential processes, the first one is the dissolution of aluminum electrodes and formation of flocs due to the polymerization of aluminum oxy-hydroxides as shown in the following equations:



While the second essential process is the simultaneous production of hydrogen at the cathode electrode [11] as shown in equation (3).

Different hydroxo-complexes including monomers and polymers will be formed such as $\text{Al}(\text{OH})^{+2}$, $\text{Al}(\text{OH})_2^+$, $\text{Al}_2(\text{OH})_2^{+4}$, $\text{Al}_6(\text{OH})_{15}^{+3}$, $\text{Al}_7(\text{OH})_{17}^{+4}$, $\text{Al}_8(\text{OH})_{20}^{+7}$, $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{+7}$, and $\text{Al}_{13}(\text{OH})_{34}^{+5}$ [12].



Response surface methodology, or (RSM), is classified as a collection of mathematical and statistical useful techniques for analyzing how far these electrodes are consumed during this technique of wastewaters treatment where several independent parameters influence a dependent response [13].

Experimental work

1. Apparatus

The electrocoagulation cell with a volume of one litre consisted of three concentric aluminium tubes with an active area equals 285 cm^2 as shown in Fig. 2 and their dimensions listed in Table 1. Additional tools were used to complete that unit and other used to measure the required information as follows:

- Digital DC- power supply (SYADGONG company-305D); 0-30 volt and 0-5 Ampere.
- Digital balance (500g x 0.01g) (PROF company).
- Magnetic Stirrer (ALFA company: HS-860) ; 0-1000 rpm.
- pH meter (ATC company).
- Digital timer (SEWAN company).

Table : Electrodes dimensions.

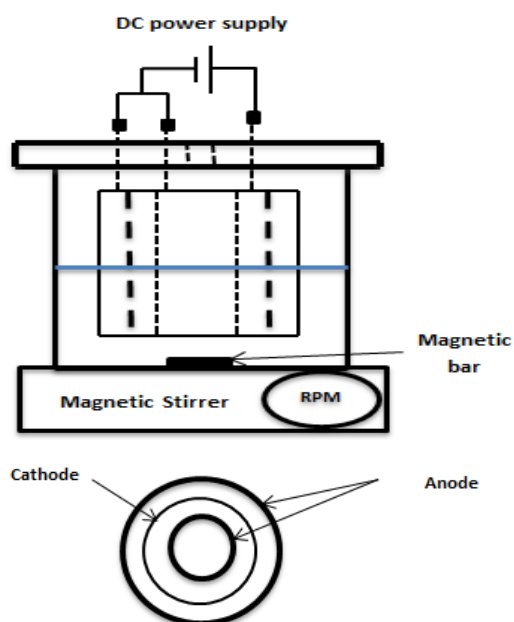
Electrode Location	Electrode thickness (cm)	Outer diameter (cm)	Inner diameter (cm)
Outer electrode	0.20	7.50	7.30
Mid. electrode	0.15	5.70	5.55
Inner electrode	0.30	4.00	3.70

2. Experimental procedure

Samples of synthesis wastewater with 155ppm of an initial concentration of lead were prepared by dissolving ion nitrate $Pb(NO_3)_2$ having 99.99 of purity (B.D.H- England) in 500mL of distilled water.

Hydrochloric acid (0.1 N) and sodium hydroxide (0.1 N) was used to adjust the value of solution pH to be as designed.

The procedure of this process occurs when the electrodes of the concentric tubes immersed in the synthesis wastewater, DC- current switched on to supply current to the cell. The interval collected samples are filtered by the cellulose Glass-Microfibre discs (Grade: MGC; pore diameter is 0.47 micrometer- MUNKTELL) before the analysis by the Atomic Absorption Spectroscopy (AAS-Type- SHIMADZUAA -7000F) for measuring quantities of lead ions removed from the solution which equals the value of aluminium ions released from electrodes during the period of each experiment.


Figure 2: Schematic of the electrocoagulation cell and the electrodes configuration.

At the end of each experiment, electrodes were washed one time with HCl and more than one time with distillate water to ensure it was cleaned well. Then, the same procedure was repeated for the next experiment.

The studied operating parameters that may affect the degree of electrodes consumption are listed in Table 2 as follows:

Table 2: Operating variables values.

Parameters	The value
Initial lead concentration (ppm)	155
pH	2-12
Current or current density (A or mA/cm ²)	0.2-2.6 or (0.702-9.123)
Stirring speed (RPM)	150
Contact time (min.)	5 - 60

Results and discussion

The classification of electrodes consumption is not clear to be a useful point or such as a drawback. Therefore, the analyzing of this objective should be taken into consideration to complete the understanding of how electrocoagulation is worked.

Experiments are designed according to Box-Wilson method where the method of least squares is used to estimate the coefficients values in the used model from experimental data to get the solution of an equation. The unknown coefficients are estimated by using the STATISTICA-10 and MINITAB-17 software programs.

The following results explained how far each one of the operating parameters affecting the required responses.

1. Anode consumption

Theoretical anode consumption (TAC) or the amount of anode material dissolved is determined according to the following equation [3]:

$$TAC (g) = I \cdot t \cdot M / Z \cdot F \quad (8)$$

Where: **I** is the current in (Amps.), **t** is the electrolysis time in (second), **M** is molecular weight in (g/mol.), **Z** is the number of electrons involved in the reaction (for Al is 3), and **F** is Faraday's constant which equals (96485.34 Columb/mol.).

Figure 3 shows the direct relation between the consumed value of anode electrode and the period of the experiment belong to the next correlation:

$$TAC (g) = 5.013 \cdot 10^{-16} + 0.0084 t + 4.289 \cdot 10^{-19} t^2 \quad (9)$$

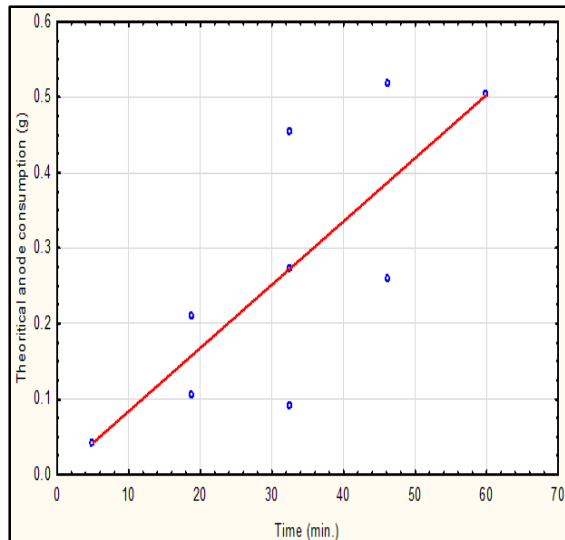


Figure 3: Theoretical anode consumption vs. time at mean values of pH and applied current.

The actual anode consumption is measured practically by weighting this electrode after each experiment. Figure 4 explains the direct relation between the consumed value of anode electrode and the duration of experiments when other operating variables are taken to be at their mean values.

Moreover, Fig. 5 indicates an increase in the consumption of anode when pH of the synthesis solution is increased until it reaches approximately 9.5 and then decreases when pH increases to more than 9.5 due to the controlled quantities of flocs which are produced by the aluminum ions released as a result of the supplied electric current.

The important effect of the direct current supplied to the electrocoagulation cell is presented clearly in Fig. 6 which caused the continuous releasing of Al(III) and consumption of anode electrode as a consequent.

The continuous releasing of hydrogen at cathode electrode as mentioned in equation (3) caused the change of pH value, Fig. 7 shows the relation between the actual anode consumption and the final pH of the simulated wastewater.

The mathematical correlations that relate the actual anode consumption with time, pH, current, and final pH are listed in Table 3 as follows:

Table 3: Mathematical correlations of the actual anode consumption.

Operating variable (x)	Correlation of the actual anode consumption (Y)
Time (min.)	$Y = 0.0743 + 0.0031 x + 0.0001 x^2$
pH	$Y = -0.0109 + 0.0888 x - 0.0056 x^2$
Current (Amps.)	$Y = -0.0128 + 0.2782 x - 0.0397 x^2$
Final pH	$Y = -1.2848 + 0.3827 x - 0.0225 x^2$

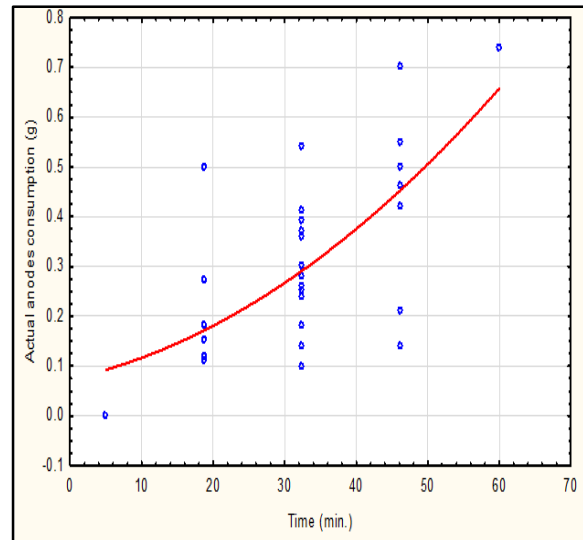


Figure 4: Actual anode consumption vs. time at mean values of pH and applied current.

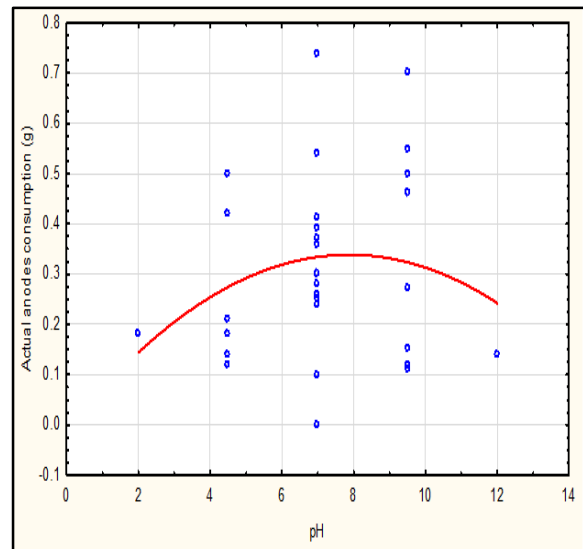


Figure 5: Actual anode consumption vs. pH at mean values of time and applied current.

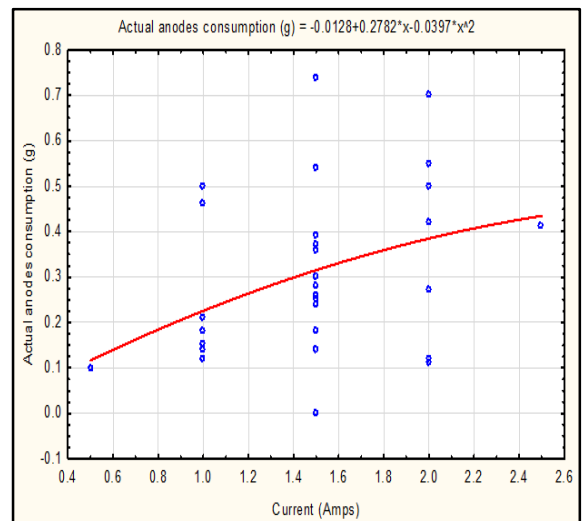


Figure 6: Actual anode consumption vs. current at mean values of time and pH.

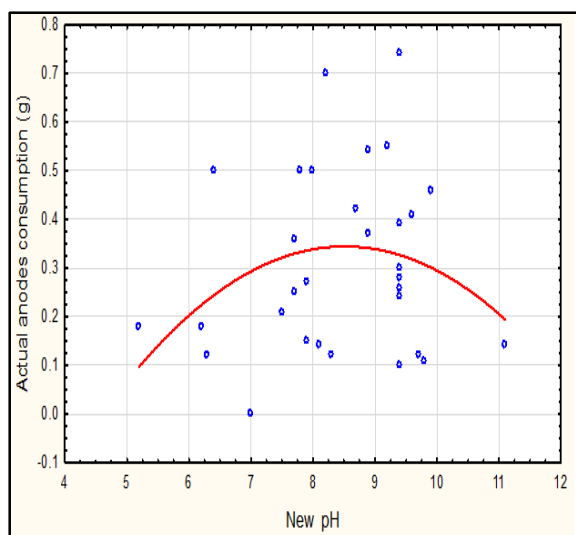


Figure 7: Actual anode consumption vs. final pH at mean values of time and applied current.

Figure 8 shows plainly how the value of solution pH is affecting the value of anode consumption. This value is higher when pH is low while it increased along the period of the experiment when the value of pH be larger. Table 4 listed the correlations of Fig. 8 and their coefficients of determination R^2 .

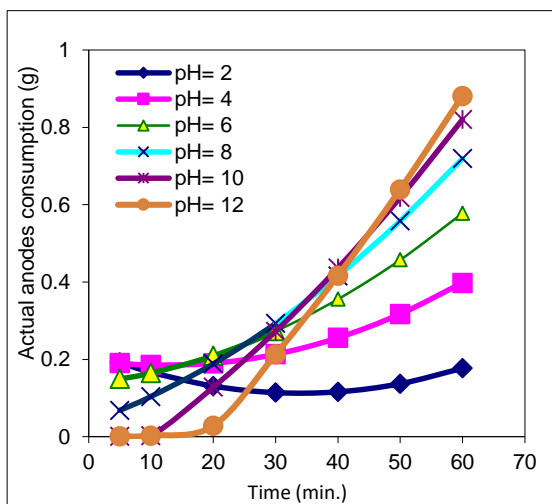


Figure 8: Actual anode consumption vs. time with several constant pH at mean value of applied current.

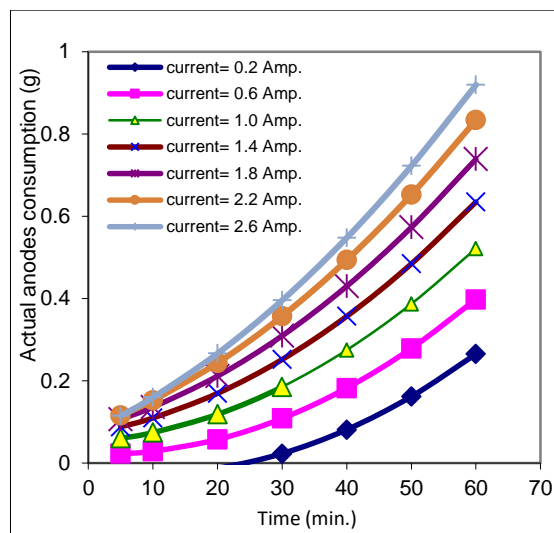


Figure 9: Actual anode consumption vs. time with several constant current at mean value of pH

Table 4. Mathematical correlations of the actual anode consumption vs. time with several constant pH at mean value of applied current

pH	Actual anodes consumption correlation	R^2
2	$y = 1E-04x^2 - 0.0064x + 0.2219$	1.000
4	$y = 9E-05x^2 - 0.0024x + 0.2004$	1.000
6	$y = 9E-05x^2 + 0.0016x + 0.1389$	1.000
8	$y = 9E-05x^2 + 0.0057x + 0.0377$	1.000
10	$y = 0.0001x^2 + 0.0071x - 0.0575$	0.9982
12	$y = 0.0003x^2 + 0.0003x - 0.0270$	0.9936

Table 5: Mathematical correlations of the actual anode consumption vs. time with several constant current at mean value of pH.

Current (Ampere)	Actual anodes consumption correlation	R^2
0.20	$y = 9E-05x^2 + 6E-05x + 0.0116$	1.000
0.60	$y = 9E-05x^2 + 0.001x + 0.0376$	1.000
1.00	$y = 9E-05x^2 + 0.0018x + 0.0598$	1.000
1.40	$y = 9E-05x^2 + 0.0027x + 0.0784$	1.000
1.80	$y = 1E-04x^2 + 0.0036x + 0.0933$	1.000
2.20	$y = 9E-05x^2 + 0.0054x + 0.1120$	1.000
2.60	$y = 9E-05x^2 + 0.0072x + 0.1160$	1.000

Figure 9 explains clearly the directly affecting of applied current on the rising of the anode consumption value and continuous releasing of aluminum ions along the periods of the experiments for all of the current values supplied from the DC-supplier. Curves correlations and their R^2 are listed in Table 5.

Practicality, the actual anode consumption is larger than the theoretical values but their direct relation is clear as shown in Fig. 10.

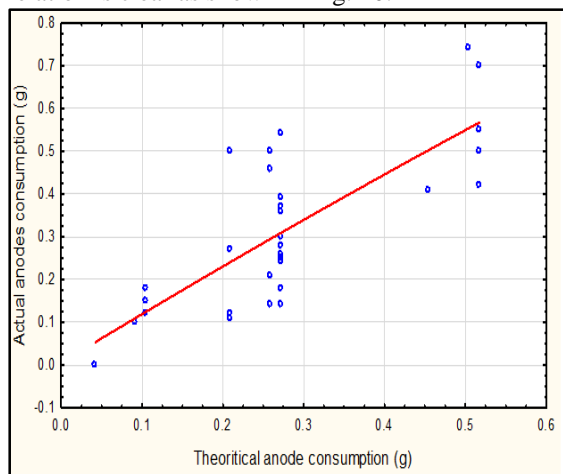


Figure 10: Actual anode consumption vs. theoretical anode consumption.

The correlation that relates actual anode consumption (AAC) with theoretical consumption (TAC) is shown in equation (10) as follows:

$$AAC (g) = 0.0052 + 1.1531 (TAC) - 0.1284 (TAC)^2 \quad (10)$$

2. Cathode consumption

As mentioned before that there is also a consumption operation on the cathode aluminum electrode [7]. Therefore the same procedure of anode consumption analysis will be doing to analysis the consumption of cathode.

Table 6 presented the relations between the cathode consumption and the studied operating parameters. Figures 11 to 14 show the variation of cathode weight consumed along each of operating parameters when others are at their mean values.

Moreover, Fig. 15 explains the affecting of the variation of the solution pH value on the amounts of cathode electrode consumed along the duration of each experiment. Table 7 listed curves correlations and their coefficients of determination.

While Fig. 16 shows the affecting of current supplied to electrocoagulation cell on the value of cathode consumed along the period of each of the treatment experiments where their correlations and R^2 are listed in Table 8.

Figures 17 to 20 show in double Y-axis plots the consumption of anode and cathode electrodes vs. time, pH, applied current, and final pH respectively.

Table 6: Mathematical correlations of the cathode consumption.

Operating variable (x)	Correlation of the actual anode consumption (Y)
Time (min.)	$Y = 0.0365 + 0.0015 x + 7.6253E-5 x^2$
pH	$Y = 0.1287 + 0.0153 x - 0.0011 x^2$
Current (Amps.)	$Y = 0.2647 - 0.1785 x + 0.0737 x^2$
Final pH	$Y = -0.8239 + 0.2422 x - 0.0143 x^2$

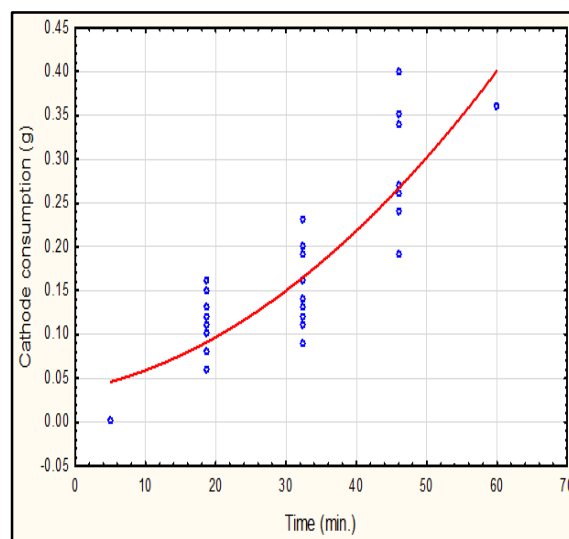


Figure 11: Cathode consumption vs. time at mean values of pH and applied current.

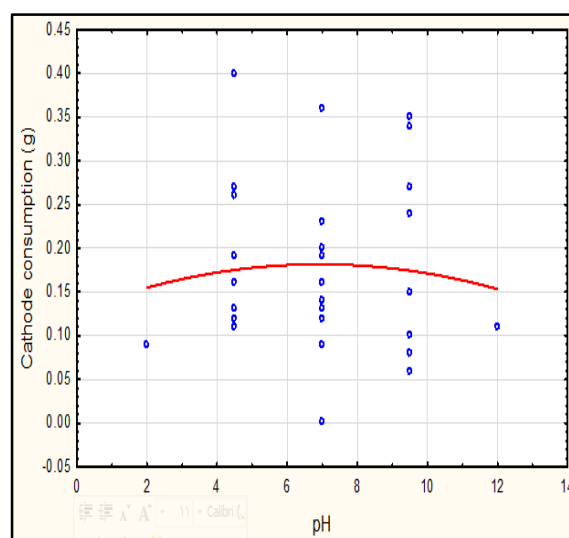


Figure 12: Cathode consumption vs. pH at mean values of time and applied current.

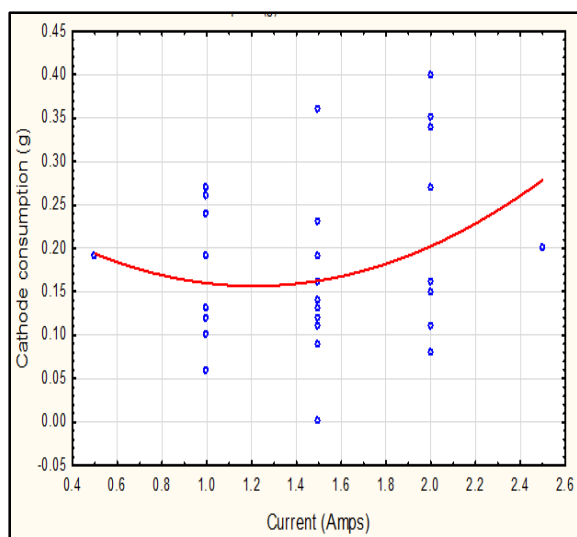


Figure 13: Cathode consumption vs. current at mean values of time and pH.

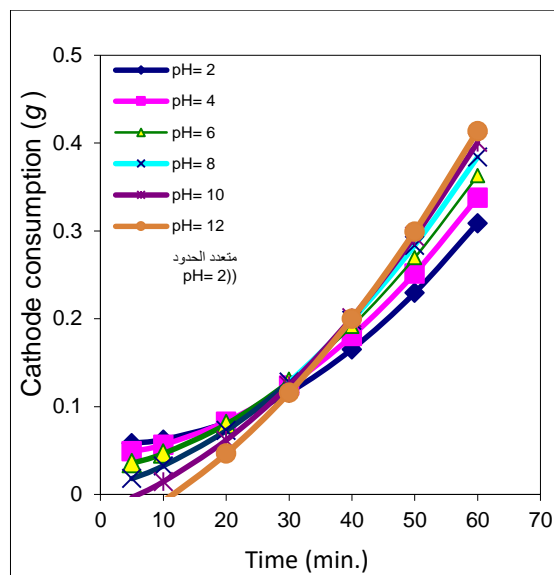


Figure 15: Cathode consumption vs. time with several constant pH at mean value of applied current.

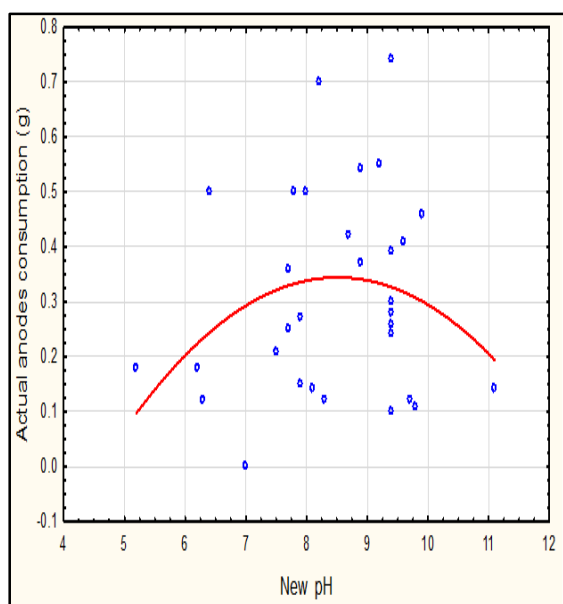


Figure 14: Cathode consumption vs. final pH at mean values of time and applied current.

Table 7: Mathematical correlations of the cathode consumption vs. time with several constant pH at mean value of applied current.

pH	Actual anodes consumption correlation	R ²
2	$y = 7E-05x^2 - 0.0003x + 0.0581$	1.000
4	$y = 8E-05x^2 + 0.0004x + 0.0453$	1.000
6	$y = 7E-05x^2 + 0.0011x + 0.0283$	1.000
8	$y = 7E-05x^2 + 0.0018x + 0.0073$	1.000
10	$y = 7E-05x^2 + 0.0025x - 0.0178$	1.000
12	$y = 7E-05x^2 - 0.0003x + 0.0581$	1.000

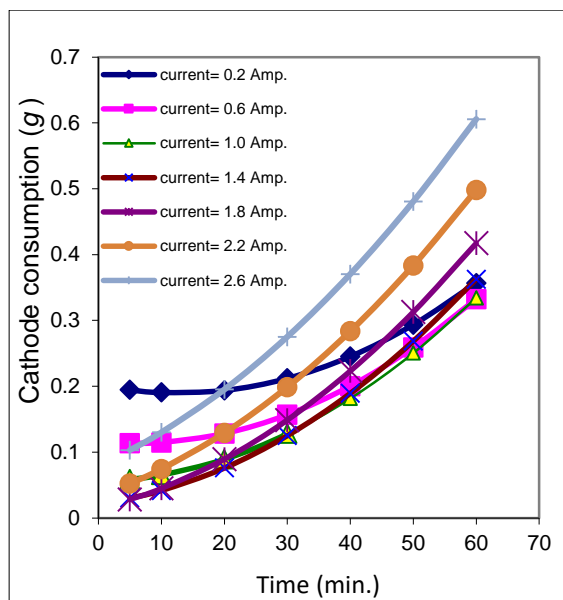


Figure 16: Cathode consumption vs. time with several constant current at mean value of pH.

Table : Mathematical correlations of the cathode consumption vs. time with several constant current at mean value of pH.

Current (Ampere)	Actual anodes consumption correlation	R ²
0.20	$y = 8E-05x^2 - 0.0019x + 0.2025$	1.000
0.60	$y = 7E-05x^2 - 0.0009x + 0.1162$	1.000
1.00	$y = 8E-05x^2 + 0.0001x + 0.0562$	1.000
1.40	$y = 8E-05x^2 + 0.0012x + 0.0226$	1.000
1.80	$y = 7E-05x^2 + 0.0022x + 0.0153$	1.000
2.20	$y = 7E-05x^2 + 0.0032x + 0.0344$	1.000
2.60	$y = 8E-05x^2 + 0.0043x + 0.0798$	1.000

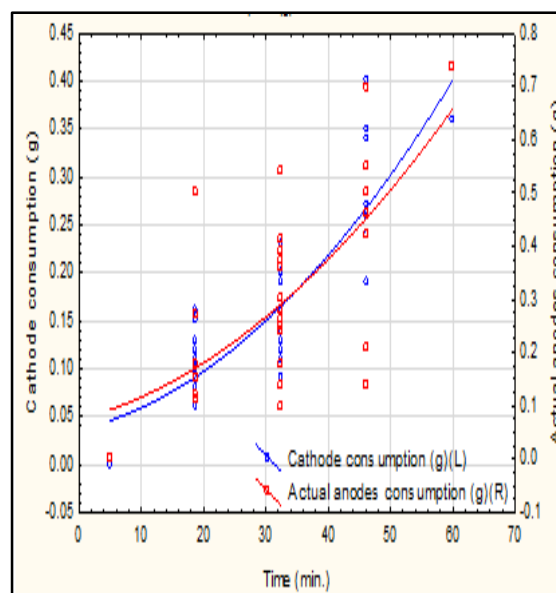


Figure 17: Cathode and actual anode electrodes consumption vs. time at mean values of pH and applied current.

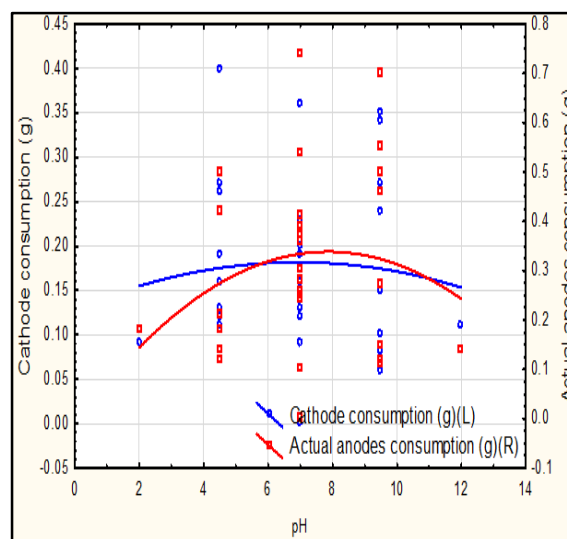


Figure 18: Cathode and actual anode electrodes consumption vs. pH at mean values of time and applied current.

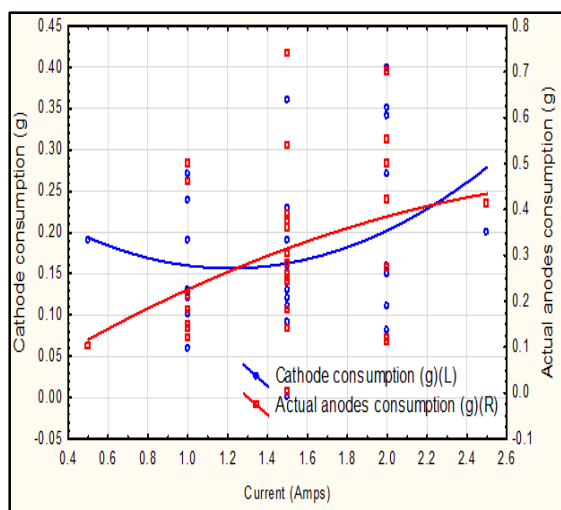


Figure 19. Cathode and actual anode electrodes consumption vs. applied current at mean values of time and pH

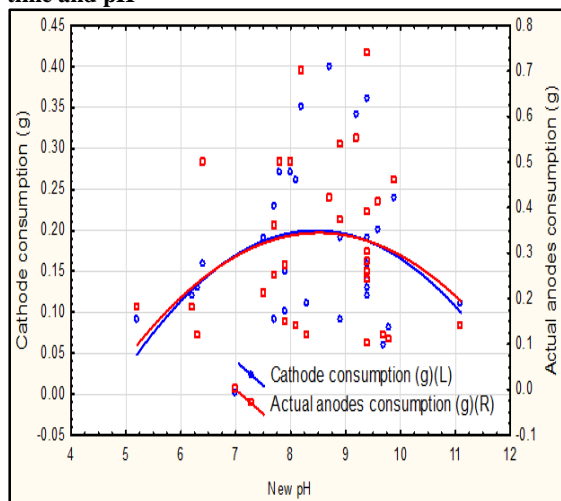


Figure 20: Cathode and actual anode electrodes consumption vs. final pH at mean values of time and applied current.

Conclusions

The present study has shown that consumption occurs in both aluminium electrodes (i.e. cathode and anode).

Moreover, it proved that the actual anode consumption is higher than its theoretical value.

The studied operational variables have significant effects on the consumption of electrodes, especially the applied electric current.

The effect of the pH value on the amount of consumption of electrodes varies over the duration of each experiment depending on its value change over time.

Several mathematical correlations were obtained which relate between the dependent and independent variables and which may be useful in the simulation analysis for the scaling up of this technique of wastewater treatment.

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