

# Optimization of copper and nickel removal by electrocoagulation through a recirculation flow bipolar reactor: statistical modeling

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ARTICLE INFO	ABSTRACT/RESUME				
Article History :	Abstract: Heavy metals have a detrimental effect on human health				
Received : 04/05/2021 Accepted : 17/01/2022	and environment if not managed efficiently. For this reason, industrial wastewaters are strictly regulated and have to be treated before being discharged. The efficiency of bipolar reactor with				
Key Words:	recirculation flow mode and aluminum electrode in removing copper				
Electrocoagulation; Heavy metal removal; Statisticalmodeling; Optimization.	and nickel from high concentrated solution by electrocoagulation was examined. The experimental design (Box-Behnken) was employed to investigate the effects of different operating conditions and their interactions on the removal: electrolysis time (t, min), current density (i, $A/m^2$ ), pH, and initial concentration ( $C_i$ , mg/L).				
	The results showed a high removal rate (97.42% for copper, and 100% for nickel), and also a good correlation between the values measured and those predicted by the Box–Behnken experimental design, for the removal of copper ( $R^2 = 0.94$ , P value = 0.0003) as well as of nickel ( $R^2 = 0.95$ , P value = 0.0001).				

## I. Introduction

The presence of heavy metals in industrial wastewater discharge by many industries of developed countries, and the rejection of wastewater without treatment, increases the contamination of our environment resulting in affecting human health in the short and the long term [1].

Various techniques have been employed for the treatment of heavy metals, including precipitation [2], adsorption [3, 4], biosorption [5], ion-exchange [6], electro-dialysis [7], and membrane separation [8, 9]. Several studies have been carried out to improve treatment processes of heavy metal and reduce their costs [10-12]. The electrocoagulation processes has proven by multiple researchers for capability of heavy metal removal from all kind of polluted water [13, 10], research has been continuing in recent years in improving the performance of this method in terms of the reactor [14, 12] and the many variables that affect its performance.

The electrocoagulation processes consisting of creating metallic hydroxide flocks by electro-dissolution of soluble anodes (Aluminum, or iron ...), the generation of metallic cation, and the hydroxide metal formed could adsorb or neutralize the polluted particles. These neutralize particles agglomerate to form flocs that are susceptible to decantation or flotation [15, 16].

The objective of this research was to study the removal of high copper and nickel concentrations from aqueous solution by the EC process, using recirculation flow bipolar reactor with Al electrodes, the recirculation flow was used to improve the performance of the treatment. The response surface methodology (RSM) was used to give a statistical modeling making possible to describe the effects and the relationships of the four operating parameters such as pH, current density, residence time and initial concentration.

# **II. Experimental**

# **II.1.** Experimental setup

The reactor used in this study is a bipolar electrocoagulator with recirculation flow mode. It consists of a parallelepiped shaped plexiglas cell. The synthetic effluent is pumped into the electrochemical cell containing three aluminum plates used as electrodes (Dimensions  $20 \times 10$  cm) and deposited vertically in the reactor (size  $7 \times 10 \times 20$  cm). The gaps between the two adjacent electrodes were kept constant at 1 cm for all the experiments. The electrochemical cell has a volume of about 1.025 L. Schematic diagram of the An agitator has been put in place in which the effluent is homogenized, to prevent the formation of foams

experimental setup shown in Figure 1. which can disturb the reactions and the flow of the effluent and prevent the proper functioning of the EC



Figure 1. Installation of the experimental devices

## II.2. Experimental procedure and analytic methods

A synthetic solution was prepared by dissolution of nickel sulfate or copper sulfate with sodium chloride in tap water. NaCl (1g/L) plays the role of supporting electrolyte

during the experiment, 2L of the solution was treated through 1L of solution containing in the reactor, and that is done by a recirculation flow, the samples are taken at defined times (residence time), 100 ml are taken in a beaker and after 30 minutes of decantation, 10 ml of the solution are taken with a syringe.

In this study, we used aluminum electrodes [17, 18]. These electrodes are immersed in the solution. One of the factors that affect the efficiency of the process is the state of the electrodes, so before each test, they must be cleaned to avoid all kinds of impurities: rinsing with distilled water; rinsing with the NaOH solution (10 %); and rinsing with tap water.

The pH of solution was adjusted to the desired values by either 0.1 M HCl or 0.1 M NaOH; the measurements were carried out with a pH meter (HANNA pH 301) fitted with a combined glass electrode (HANNA Hi 1131B). The device is calibrated with standard buffer solutions (pH = 4, 7 and 10). The measurement of the concentration of metal ions in solution is carried out by atomic absorption spectrophotometer SAA model ANALYTIK JENA AG GERMANY, AAS NOVAA 350.

## II.3. Box-Behnken design

Box-Behnken designs are used to generate higher order response surfaces using fewer required runs than a normal factorial technique, it has been proven that is a good design by many researchers, for the optimization of water treatment [19, 20], and it permits, estimation of the parameters of the quadratic model; building of sequential designs, detection of lack of fit of the model [21].

In this research, the Box–Behnken experimental design is applied to give us a statistical modeling to examine the treatment process parameters affecting the removal of metal by electrocoagulation. Residence time  $(X_1)$ , current density  $(X_2)$ , pH  $(X_3)$  and initial concentration  $(X_4)$  are input variable parameters. The factor levels were coded as -1 (low), 0 (central point or middle) and 1 (high).

The rate of metal removal, Y (%) was chosen as a response of the studied system and it was calculated by the following equation:

$$Y(\%) = \left[\frac{[M]_i - [M]_f}{[M]_i}\right] \times 100$$
(1)

Where  $[M]_i$  and  $[M]_f$  are the concentration of the metal before and after the treatment, respectively.

For this response (Y), a polynomial model of the second degree is established to quantify the influence of the variables,

$$Y_{i} = \beta_{0} + \sum_{1}^{4} \beta_{i} X_{i} + \sum_{1}^{4} \beta_{ii} X_{ii}^{2} + \sum_{1}^{4} \beta_{ij} X_{ij}$$
(2)

Where  $\beta_0$ ,  $\beta_i$ ,  $\beta_{ii}$  and  $\beta_{ij}$  are the regression coefficients for intercept, linear, quadratic and interaction terms, respectively, and *Xi*, and *Xj* are the independent variables



<b>Tuble 1.</b> Experimental design levels of chosen variables							
Variables	Levels in Box–Behnken design						
Coded level	Low (-1)	Middle (0)	High (+1)				
$(X_I)$ Residence Time, $t$ (min)	10	25	40				
( $X_2$ ) Current density, $i$ (A/m <sup>2</sup> )	40	80	120				
$(X_3)$ Initial pH (pHi)	4	7	10				
( $X_4$ ) Initial concentration, $C_i$ (mg/L)	30	60	90				

**Table 1.** Experimental design levels of chosen variables

## III. Results and discussions

## **III.1. Statistical analysis**

The combined effects of residence time, current density, pHi and initial concentration multilevel for metal removal were monitored. Table 2 shows the data

resulting from the experiments of the effect of the four variables on the treatment. The experimental results were analyzed through an R S M design by using JMP Pro 13 program. The predicted results by the Box-Behnken design are shown in Table 2.

Table 2. Coded and actual levels with experimental and predictive percentage removal values of copper and nickel

N.	Code	d level	of varia	ables	A	Actual level of variables			percentage removal Y %			
exp	$X_{I}$	$X_2$	$X_3$	$X_4$	t	i	pHi	$C_i(mg/L)$	(	Cu	Ν	Ji
					(min)	$(A/m^2)$			Exp	Pred	Exp	Pred
1	-1	-1	0	0	10	40	7	60	72.38	67.75	58.63	62.01
2	-1	0	1	0	10	80	10	60	94.20	100.62	85.22	82.18
3	0	-1	0	-1	25	40	7	30	89.51	88.95	97.86	91.43
4	-1	0	0	1	10	80	7	90	79.92	71.13	70.86	69.63
5	1	0	0	1	40	80	7	90	96.23	95.76	98.45	99.00
6	1	0	0	-1	40	80	7	30	89.98	95.53	99.35	98.89
7	0	0	1	1	25	80	10	90	96.03	97.81	100	101.06
8	0	-1	-1	0	25	40	4	60	48.99	49.02	88.05	85.26
9	0	1	0	-1	25	120	7	30	97.42	97.15	100	101.37
10	1	0	1	0	40	80	10	60	94.41	86.91	100	97.53
11	1	1	0	0	40	120	7	60	95.68	101.72	100	96.98
12	0	0	0	0	25	80	7	60	91.25	92.63	99.49	99.43
13	0	0	-1	1	25	80	4	90	52.63	55.66	96.46	93.35
14	0	1	-1	0	25	120	4	60	93.68	88.37	100	98.40
15	0	-1	0	1	25	40	7	90	87.55	88.95	86.55	91.43
16	0	1	1	0	25	120	10	60	95.85	93.27	100	101.08
17	0	-1	1	0	25	40	10	60	95.85	98.62	95.83	95.72
18	0	0	1	-1	25	80	10	30	91.27	90.34	100	103.45
19	1	0	-1	0	40	80	4	60	94.97	90.35	97	101.33
20	1	-1	0	0	40	40	7	60	95.23	96.20	93.33	94.36
21	-1	1	0	0	10	120	7	60	92.89	93.33	77.38	76.71
22	0	0	-1	-1	25	80	4	30	77.55	75.10	97.48	96.86
23	0	1	0	1	25	120	7	90	96.87	98.52	94.32	97.13
24	-1	0	-1	0	10	80	4	60	30.50	39.80	60.30	64.06
25	-1	0	0	-1	10	80	7	30	86.09	83.32	77.88	75.64

The plot of experimental and predictive values of *Y* are shown in Figure 2, the  $R^2$  and *Pvalue* indicate that the data correlated fairly well for the copper removal ( $R^2$  =

0.94, *P* value = 0.0003) as well as the nickel removal  $(R^2 = 0.95, P \text{ value} = 0.0001).$ 



Figure 2. Actual by predicted plot

The coefficients values of the parameters are listed in Table 3, the *Pvalue* are used as a tool to check the significance of each coefficient, which in turn, can indicate to the variables and their interactions.

The *Pvalue* of  $X_1 * X_3$  (effect of interaction between pH and electrolysis time) is a lower than the other factor, that indicate the high influence on the electrocoagulation process for copper removal, and for nickel removal the electrolysis time has the most important significance than other factor, as shown in Table 3.

Table 3	8. Effect	summary
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Copper								
Source	LogWorth	Pvalue						
$X_{1}(t)*X_{3}(pHi)$	3.137	0.00073						
$X_3$ (pHi)	2.931	0.00117						
$X_{l}$ (t)	2.129	0.00744						
$X_2(i) * X_3(pHi)$	2.065	0.00861						
<i>X</i> <sub>3</sub> (pHi)* <i>X</i> <sub>3</sub> (pHi)	1.546	0.02844						
$X_3 (\text{pHi}) * X_4 (Ci)$	1.183	0.06568						
$X_2(i)$	0.992	0.10183						
$X_{1}(t)^{*}X_{2}(i)$	0.782	0.16537						
<i>X</i> <sub>2</sub> (i)*X <sub>4</sub> (Ci)	0.520	0.30201						
$X_4$ (Ci)	0.463	0.34459						
$X_{1}(t)*X_{4}(Ci)$	0.425	0.37594						
$X_{l}(t)^{*}Xl(t)$	0.356	0.44025						
$X_4(Ci)^*X_4(Ci)$	0.332	0.46508						
$X_{2}(i)^{*}X_{2}(i)$	0.035	0.92293						

Nickel							
	1						
Source	LogWorth	Pvalue					
$X_{l}(t)$	3.942	0.00011					
$X_{l}(t)^{*}X_{l}(t)$	3.262	0.00055					
$X_{1}(t)*X_{3}(\text{pHi})$	1.528	0.02965					
$X_{2}(i)$	1.505	0.03126					
$X_2(i) * X_2(i)$	0.866	0.13619					
$X_{1}(t)^{*}X_{2}(i)$	0.715	0.19273					
$X_3$ (pHi)	0.604	0.24903					
$X_2(i) * X_3$ (pHi)	0.438	0.36459					
$X_{1}(t)$ * $X_{4}(Ci)$	0.305	0.49536					
$X_2(i) * X_4(Ci)$	0.111	0.77419					
$X_4 (Ci) * X_4 (Ci)$	0.092	0.80988					
$X_3$ (pHi)* $X_4$ (Ci)	0.048	0.89632					
X <sub>4</sub> (Ci)	0.025	0.94318					
<i>X</i> <sub>3</sub> (pHi)* <i>X</i> <sub>3</sub> (pHi)	0.018	0.95830					

The estimated values of the parameters are reported in Table 4, for copper and nickel removal, it can be seen that the star marks indicate the high intensity of the coefficients.

Table 4. Estimated parameters

Copper								
Term	Estimate	StdError	t Ratio	Prob> t				
Intercept	-117.5649	60.3060	-1.95	0.0798				
$X_{l}\left(t ight)$	4.0902	1.2230	3.34	0.0074*				
<i>X</i> <sub>2</sub> (i)	0.8763	0.4865	1.80	0.1018				
$X_3$ (pHi)	32.6453	7.2819	4.48	0.0012*				
$X_4(Ci)$	-0.6480	0.6532	-0.99	0.3446				
$X_1 * X_2$	-0.0083	0.0055	-1.50	0.1654				
$X_1 * X_3$	-0.3569	0.0744	-4.79	0.0007*				
$X_2 * X_3$	-0.0991	0.0304	-3.26	0.0086*				
$X_1 * X_4$	0.0069	0.0074	0.93	0.3759				
$X_2 * X_4$	0.0030	0.0028	1.09	0.3020				
$X_3 * X_4$	0.0747	0.0361	2.07	0.0657				
$X_I * X_I$	-0.0144	0.0179	-0.80	0.4403				
$X_2 * X_2$	0.0002	0.0023	0.10	0.9229				
$X_3 * X_3$	-1.1065	0.4324	-2.56	0.0284*				
$X_4 * X_4$	-0.0032	0.0043	-0.76	0.4651				
		Nickel						
Term	Estimate	StdError	t Ratio	Prob>/t/				
Intercept	-29.4703	38.9143	-0.76	0.4663				
$X_{l}\left(t ight)$	4.8222	0.7892	6.11	0.0001*				
$X_{2}\left(i ight)$	0.7859	0.3139	2.50	0.0313*				
$X_3$ (pHi)	5.7511	4.6988	1.22	0.2490				
$X_4(Ci)$	-0.0308	0.4215	-0.07	0.9432				
$X_1 * X_2$	-0.0050	0.0036	-1.40	0.1927				
$X_1 * X_3$	-0.1217	0.0480	-2.53	0.0296*				
$X_2 * X_3$	-0.0186	0.0196	-0.95	0.3646				
$X_1 * X_4$	0.0034	0.0048	0.71	0.4954				
$X_2 * X_4$	-0.0005	0.0018	-0.29	0.7742				
$X_{3}^{*}X_{4}$	0.0031	0.0233	0.13	0.8963				
$X_I * X_I$	-0.0578	0.0116	-4.99	0.0005*				
$X_2 * X_2$	-0.0024	0.0015	-1.62	0.1362				
$X_{3}^{*}X_{3}$	-0.0149	0.2790	-0.05	0.9583				
$X_4 * X_4$	-0.0006	0.0027	-0.25	0.8099				

\* High intensity of the coefficients significance



The mathematical expression of the metal ion removal relationship with the four variables ( $X_1$ ,  $X_2$ ,  $X_3$  and  $X_4$ ) is shown below in terms of actual factors. Equation (3) and (4) predict the removal rate:

$$Y(\%)(Cu) = -117.5649 + 4,0902X_1 + 0.8763X_2 + 32.6453X_3 - 0.6480X_4 - 0.0083X_1X_2 - 0.3569X_1X_3 + 0.0069X_1X_4 - 0.0991X_2X_3 + 0.0030X_2X_4 + 0.0747X_3X_4 - 0.0144X_1^2 + 0.0002X_2^2 - 1.1065X_3^2 - 0.0032X_4^2$$
(3)

$$Y(\%)(Ni) = -29.4703 + 4.8222X_1 + 0.7859X_2 + 5.7511X_3 - 0.0308X_4 - 0.0050X_1X_2 - 0.1217X_1X_3 + 0.0034X_1X_4 - 0.0186X_2X_3 - 0.0005X_2X_4 + 0.0031X_3X_4 - 0.0578X_1^2 - 0.0024X_2^2 - 0.0149X_3^2 - 0.0006X_4^2$$
(4)

Table 5, shows the variance analysis (ANOVA) results for the response Y(%). The analysis of variance of the response showed that the model is highly important. This importance is shown by the value of Prob > F.

The *F* value is the ratio of the mean regression sum of squares divided by the mean error sum of the squares. The value of Prob>F is the probability that the null hypothesis for the full model is true. This low value would imply that at least some of the regression parameters are non-zero and that the regression equation does have some validity in fitting the data. The low probability value (<0.001), indicates that the model is considered statistically significant [22].

<b><i>ubic</i> 5.</b> <i>maiysis of variance</i>	Table	5.	Anal	ysis	of	variance
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Copper								
Source	DF	Sum of Squares	Mean	F Ratio				
			Square					
Model	14	6639.0118	474.215	10.5585				
Error	10	449.1329	44.913	Prob > F				
C. Total	24	7088.1447		0.0003*				
Nickel								
Source	DF	Sum of Squares	Mean	F Ratio				
			Square					
Model	14	3587.4000	256,243	13.7019				
Error	10	187.0130	18,701	Prob > F				
C. Total	24	3774.4131		0.0001*				

\* High intensity of the model validity

## III.2. Effect of variables on the removal rate

Form the previous observation on the signification of the parameter affecting the electrocoagulation processes for the copper and nickel removal, we decided to choose response surface and counter plots of predicted metal removal rate as a function of two combined parameters, electrolysis time and pHi ( $X_1*X_3$ ), current density and pHi ( $X_2*X_3$ ), initial concentration and pHi ( $X_4*X_3$ ). As it was described above on the signification of the parameter affecting the electrocoagulation possesses, it was observed on the response surface and counter plots, that is the improvement on the removal of metal due the increase of the value of the three parameter, electrolysis time, current density and the pHi.

#### III.2.1. Effect of electrolysis time and pHi

In an ideal EC process, the amount of coagulant produced into the solution is proportional to the reaction time and current. The theoretically dissolved mass of aluminum of the sacrificial anode can be quantified by Faraday's law.

The pHi value of the solution has a big role in the influence of the EC process, and this is due to the important role it plays in the formation of monomericaluminum hydroxide, and its effect on the precipitation of metal ions.

The effect of electrolysis time and pHi is well shown in Figures 3 and 4for copper and nickel respectively.

As shown by the curve in Figure 3, with increasing time and thepHi value, the rate of copper removal increases. With the pHi values 4, 5, 6, 7 and 8, 98.59% removal in 40 min of electrolysis time was achieved, and with pHi 9 and 10 the removal rate decreases over time, and at pHi 10, 100% removal rate is obtained in 10 min of time.

As shown in the curve of Figure 4, with increasing time and initial pHi value the rate of nickel removal increases. With the pHi values 4, 5, 6 and 7,100% removal rate was reached in 30 min up to 40 min of electrolysis time, and with pH 8, 9 and 10 the removal rate reached 100 % with 25 min of time and in this case a slight decrease in the removal rate can be observed at 40 min of time.

It is interesting to note that due to the electrolysis process, the pHi value of the solution in the EC reactor increases with the start of the process, a result similar to the work of Kumarasinghe [23]. The reactions at the electrodes when using aluminum are:

Anode: 
$$Al_{(s)} \rightarrow Al^{3+} + 3e^-$$
 (5)

Cathode: 
$$2H_20 + 2e^- \to H_{2(g)} + 20H^-$$
 (6)

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*Figure 3.* Response surface and contour plots for Copper removal vs. Electrolysis time and pHi,  $i = 80A/m^2$ , Ci = 60mg/L



Figure 4. Response surface and contour plots for Nickel removal vs. Electrolysis time and pHi,  $i = 80 \text{ A/m}^2$ , Ci = 60 mg/L



The generated amount of  $Al^{3+}$  ions increases over time, which in turn enter a reaction with  $OH^-$  to form various monomeric species such as  $Al(OH)^{2+}$ ,  $Al(OH)_{2^+}$ , and polymeric species such as  $Al_6(OH)_{15}^{3+}$ ,  $Al_7(OH)_{17}^{4+}$ ,  $Al_{13}(OH)_{34}^{5+}$ , which ultimately transform into insoluble amorphous  $Al(OH)_3$ according to a complex kinetics of polymerization/precipitation [24], depending on the reaction:

$$Al^{3+} + 3H_20 \leftrightarrow Al(OH)_{3(s)} + 3H^+ \tag{7}$$

The amorphous  $Al(OH)_3$  is responsible for the formation of flocs [25].

As with thepHi values>7 and after a certain time the removal rate decreases, this effect is higher for copper compared to nickel. The removal rate is high at the start of the process; this can be attributed to the precipitation of metal ions. And during the process,  $OH^-$  ions are consumed by  $AI^{3+}$ , which in turn can be attributed to the dissolution of copper ions due to the effect of the pH value on its precipitation.

The EC process has a certain buffering capacity due to the balance between the production and consumption of  $OH^-$ , which prevents a large change in pH. The buffer pH appears to be just above 7 [25-27].

#### III.2.2. The effect of current density

It has been established that the applied current density has an influence on the performance of the EC process via the dosage rate of the coagulant.

The effect of current density is illustrated in Figures 5 and 6, for copper and nickel respectively.

As shown in the curve of Figure 5, with increasing current density andpHi value between 4 and 8, the rate of copper removal increases. With the value of pHi 7 and 8 we have reached 100% removal after 40 min of electrolysis time, and with pHi 9 and 10 the removal rate decreases with time and at pHi 10 we obtain 100% of removal rate at only10 min of time.

And for nickel (Figure 6), it was observed that the removalrate increases with the increase of the current density and thepHi value without any exception where 100% removal was reached with 60, 70 and 80  $A/m^2$  for pHi 10, 9, 8 respectively.

The removal rate of copper and nickel increases with increasing of the current density [28] slightly higher for copper than for nickel. The increased removal is due to an increased flux of Al hydroxide ions required to form the coagulant [29].



Figure 5. Response surface and contour plots for copper removal vs. Current density and pHi, t = 25min, Ci = 60m/L

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Figure 6. Response surface and contour plots for nickel removal vs. Current density and pHi, t = 25min, Ci = 60mg/L

#### III.2.3. The effect of initial concentration

The effect of the initial concentration is studied and the results are illustrated in Figures 7 and 8, for copper and nickel respectively.

The curve in Figure 7, shows that as the initial copper concentration increases, the removal rate decreases at pHi<8, and for pHi>8 there is almost no effect of increasing the initial concentration on the removal rate.



Figure 7. Response surface and contour plots for copper removal vs. Init. Concentration and pHi, t = 25min,  $i = 80A/m^2$ 



It can be noticed in the curve of Figure 8, that the increase in the initial concentration of nickel decreases the removal rate. Similar results have been found by Nwabanne [30] for the removal of copper, nickel and chromium and by Kaur and Vatta [17] for the removal of arsenic and chromium.

This may be because at constant time, pHi and current density, the change in the production of coagulant and

 $H_2$  gas has no relationship with the change in the initial concentration of the solution. The adsorption capacity of  $Al(OH)_3$  is consumed due to the increase in the initial concentration of the solution [15].

Figure 8 show that the removal rate for nickel reaches 100 % at pHi>7.



Figure 8. Response surface and contour plots for nickel removal vs. init. Concentration and pHi, t = 25min, i = 80A/m<sup>2</sup>

## **IV. Evaluation of the electrical energyconsumption**

In the electrocoagulation process, the electrical energy consumption and the amount of electrode dissolved in the solution is a significant economic factor.

The electrical energy consumption, per unit volume of treated water, was calculated using Eq.8:

$$E = \frac{U.l.t}{V} \tag{8}$$

Where *E* is the specific energy consumption in kWh  $/m^3$ , *U* is the voltage in V, *I* is the direct current intensity in

A, *t* is the electrolysis time in hours and *V* is the volume of water treated in liter. The quantity of dissolved electrodes per unit volume of treated water was theoretically estimated using Faraday's law.

Table6 summarizes the amount of dissolved electrodes and electrical energy consumption per cubic meter of treated water and the corresponding removal rates at different current densities and treatment times. (pHi = 7, Ci = 60 mg/L).

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t	i	$m_{Al}$	E	Copper	Nickel
(min)	$(A/m^2)$	$(kg/m^3)$	(KWh/m <sup>3</sup> )	Y(%)	Y(%)
10	40	0.011	0.173	72.38	58.63
40	40	0.044	0.680	95.23	93.33
25	80	0.055	1.241	91.25	99.49
10	120	0.033	1.140	92.89	77.38
40	120	0.134	4.560	95.68	100

Table 6. Energy consumption

As expected, it is clear that increasing the current density and / or EC time increases the removal of heavy metal ions, which is associated with the increase in both electrical energy consumption and the dissolution of the electrodes.

In order to maximize the removal rate under the operating conditions of this study, it appears for nickel that the current density should not be less than 80 A/m<sup>2</sup> and the treatment time should not be less than 25 min, under these operating conditions the electrical energy consumption is minimized to 1.241 kWh/m<sup>3</sup> with a quantity of dissolved electrodes of 0.055 kg/m<sup>3</sup> (Table 6).

On the other hand for copper, it is accepted that the current density is 40  $A/m^2$  and the treatment time 40 min, to minimize the electrical energy consumption to 0.68 kWh/m<sup>3</sup> and the quantity of dissolved electrodes at 0.044 kg/m<sup>3</sup> (Table 6).

From these results, it appears that increasing the processing time increases the efficiency of removal compared to increasing the current intensity, which also minimizes the electrical energy consumption.

## V. Conclusion

In the present study, the performance of electrochemical treatment in the removal of heavy metals was investigated focusing on the influence of operating parameters, using response surface modeling, in particular the Box-Behnken design.

The results obtained revealed that the modeling of the designs of the experiments is a very suitable technique for optimizing the operating conditions of the EC. The response surface models developed in this study to predict metal removal efficiency were considered adequate. An analysis of variance showed a good coefficient of determination value ensuring a satisfactory fit of the second order regression model with the experimental data.

The results of this study indicate that EC is a very efficient process for removing metals (copper and nickel); we have reached 98.59% removal at 40 min of electrolysis time for the removal of copper with the values of pHi 4, 5, 6, 7 and 8, and at pHi 10 we obtain 100% of the elimination rate for 10 min of time.

We achieved 100% removal at 30 min electrolysis time for nickel removal. With pHi values 4, 5, 6

and 7, and with pH 8, 9 and 10 the removal rate reaches 100% with 25 min of time.

The proposed flow recirculation EC system offers lower electrical energy consumption with low metal dissolution of electrodes.

Electrocoagulation is a complex process involving many mechanisms, to achieve optimum performance and future advances in the application of this technology, more effort must be made to better understand the fundamental mechanisms of operations, improving the design of the reactor in terms of shap and effluent flow.

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