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Copper recovery from thickener overflow by electrocoagulation/flotation: optimization of response surface, modeling, and sludge study

A. Hasanzadeh-Sablouei[1] and S.M. Moosavirad[2*]

1. Department of Mining Engineering, Higher Education Complex of Zarand, Kerman, Iran
2. Department of Environment, Institute of Science and High Technology and Environmental Sciences, Graduate University of Advanced Technology, Kerman, Iran

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Abstract

The electrocoagulation/flotation process is a novel approach in mining industry that is implemented to return Cu metal to the production cycle, which improves copper recovery and reduces waste water. In this research work, the response surface methodology was applied to optimize the factors effective in Cu metal recovery and sludge volume produced from thickener overflow. To this end, the D-optimal experimental design was utilized. The influences of four independent parameters including the electrolysis time, initial pH, current density, and electrodes type were studied to investigate the initial Cu grade percentage (28%) and sludge volume produced from thickener overflow. All these parameters were found to have important effects on the Cu metal recovery and the sludge volume produced. The linear and quadratic models were utilized for the Cu grade and sludge volume, respectively. The importance of the independent variables and the interaction between them was assessed by ANOVA. The optimum operating conditions with 27.22% Cu grade were taken to be: electrolysis time: 6.5 min, initial pH: 6.7, current density: 50.2 A/m^2, and electrode type: Fe-Al. Similarly, for the produced sludge volume of 861 cm^3, the following conditions were found: electrolysis time: 15 min, initial pH: 4.1, current density: 48.7, and electrode type: Fe-Al. The outcomes underscored a practical viewpoint of electrocoagulation, known as an acceptable method for Cu recovery from mine industrials, especially in mineral processing plants.

1. Introduction

In the past two decades, production and consumption of copper have been incremented considerably in the world. Nowadays, with the increase in the exploitation of copper resources and the extension of industries, the need for this metal has increased and enhanced its value. Meanwhile, the copper recovery methods from the available and secondary sources have been considered. Regarding the industrial research works, 700,000 metric tons of Cu was reusable at a calculated content of approximately $6 billion. According to a research work carried out in 2010, about 24 million tons of copper was produced globally, 35% of which originated from recycled copper. Iran has ranked the 17th among the countries that hold copper reserves with about 5% of world copper resources. The extraction and mineral processing operations of copper have highly progressed in Iran in the recent years due to the price increments and demand for new labor abroad. This has resulted in 50% of copper utilized in the process of copper production [1].

The Miduk copper mine is extracting open pit and it is one of the largest copper mines in Iran. The concentration plant of the Miduk copper complex is located about 1.5 kilometers northeast of mine. Based on the design, 5 million metric tons per year of deposit can produce 150,000 tons of

Corresponding author: s.m.moosavirad@kgut.ac.ir (S.M. Moosavirad).

copper concentration with 30% grade. The production process of the Miduk concentration plant is considering crushing, material transfer units, material classification, flotation (Flotation air is provided by low pressure blowers for the mechanical cells and dedicated compressors for the column cells [2]), and beneficiation and dewatering. The dewatering unit is one of the most desirable places to copper recovery in production cycle.
Settling or sedimentation of solids inside a liquid by the gravity force is one of the most widely used methods of solid-liquid separation. Thickeners are major operation proceeding in metal recovery and are utilized to separate solids from liquid [3]. Therefore, equipment manufacturers and design groups have typically applied scale-up factors to design the industrial process' and equipment [4, 5]. In the mining industry, concentrate feed into the thickener supply from flotation cells. Before flotation process, the ore particles should be smaller wherever fine particles appropriate for froth flotation. Comminution and flotation are the high energy consuming stages for copper ore, since every piece of ore-regardless of grade-must be crushed, ground and processed. Froth flotation is one of the most important operation handled for the recovery of sulfide ores such as copper [6].
Flocculants are frequently applied to form a loop of solid particles and to make a more extensive aggregate to speed up sedimentation in the thickeners. The inlet slurry is distributed into two flows of overflow that possess clarification water and downstream which is an underflow of the pulp with a numerous concentration [7-9].
Figure 1 shows that in flotation cells, overflow (feed concentrate) in froth phase is discharged into the thickener. The bubbles trend to break and solids from the froth phase also toward to sediment at the bottom of the thickener. Then, Cu concentrate is treated by filter press to water recycling from Cu concentrate The ultimate production of the thickening phase is contained approximately thirty percent of copper concentrate and other metals which it is in available condition as waste material disposal into the tailing dam. Therefore, It is necessary create innovation technology to recovery of Cu metal and water extraction. In this research, for preventing from wasting water and copper recovery, is implemented electrocoagulation/flotation (ECF) process as a novel approach to return Cu metal to production cycle in concentration plant. Hence, Fine concentrate from thickener transfer to electrocoagulation reactor.

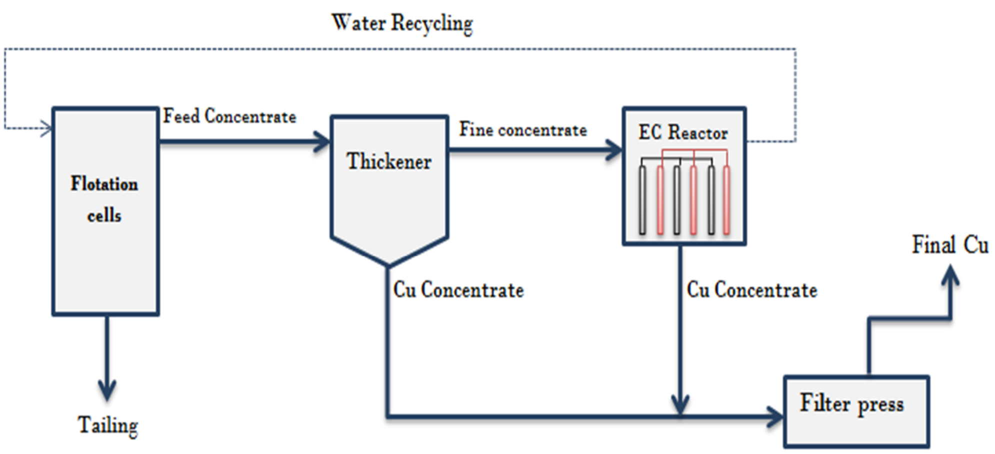

Figure 1. Experimental circuit.

The common procedures for Cu removal from water contain membranes (RO), coagulation, ion exchange, co-precipitation, electrodialysis, and adsorption [10-16]. The physical methods have demonstrated to be either too much costly or ineffective to copper removal from water for example ion exchange, electrodialysis and reverse osmosis. In the past few years, the treatment of chemical systems are not utilized because of high conservation charges, problems relevant to sludge management and effluent neutralization [17]. Ion exchange system needs resin reduction or substitution with a great expenditure when highly influence in removal of major charged metals. In other physical methods, UF and RO are clear systems which could be ample costly.
The chemical coagulation is established alkali material such as lime to increase the pH and iron

and aluminum salt to removal colloidal matter as hydroxides. However, chemical coagulants have certain disadvantages, as they require tight control over their residual concentration in treated water for human consumption as well as in industrial food production [18]. Whenever chemical precipitation is an ordinary process that produces a significantly volume of sludge [19]. With regard to expressed processes and need for low cost influence treatment, expedited many studies on the usage of electrocoagulation for the metal recovery [20, 21].

Recently, investigation on electrocoagulation system was utilized for the removal of metals, such as Cu, Cr and Ni with Fe–Fe and Al–Al electrodes from a metal plating and galvanic wastewater [22, 23].

Electrocoagulation is a simple and effective strategy wherewith flocculated kinds are produced using electro-oxidation of an anode of iron or aluminum. In this system, the separation is accomplished without producing any synthetic coagulant material or flocculent, hence, decreasing quantity of produced solid material that should be repelled of this system.

Several advantages that electrocoagulation process can be wide attractive separation is following: high particulate removal efficiency, the process is not complex since no any coagulant material to be added to water, equipment need small space, and the feasibility of full automation [24,25]. This process is distinguished by minimum sludge production and short time of operation [17, 26, 27]. However, some of the limitations are including need for maintenance, electrode passivation over time, for high efficiency need to high-conductivity water and lack of systematic reactor design [28, 29].

Electrocoagulation implicates the generation of coagulants material using dissolving electrically aluminum, iron or combination with together. In the anode, the production of metal ions be accomplished, and also in the cathode, hydrogen gas is separated from on electrode. The hydrogen gas have ability which can assist to move up the flocculated particles. In other words, the aluminum or iron hydroxide flocs ordinarily operate as adsorbents for Cu in solution. Thus, they would remove Cu from the solution [30, 31].

Accordingly, the objective of this study is to assessment on the recovery of Cu [II] ions and produced sludge volume through application of the electrocoagulation process in thickener overflow. Process parameters, namely current density, initial pH, and electrolysis time can remarkably affect the implement of electrocoagulation separation. Thus, these parameters should investigated and optimized in this research.

Response surface methodology (RSM) is considered as a significant instrument in Design Expert Software, which is largely applied for modeling and optimization of the diverse processes. In recently, RSM has been successfully occupied for treatment in dirtiest waste water such as pulp and paper, petroleum refinery, chromium, textile, electroless plating industries [32-37].

RSM is widespread admitted as a based technique of statistical in experiments design field, assessing exclusive and interplay influences of independent variables, and optimization of process indexes in condition of restricted number of experimental run [38-39]. The utilization of RSM is proved which this planning could be considerably effective for optimization and prediction the electrocoagulation processes [40-42].

The D-optimal matrix is a superb technique to investigation the effect of main parameters on the response with considering reducing the experiments number and in resulted cause to decrease consuming energy, time, and expense. With regard to our comprehension, D-optimal has not been perused and published much for the Cu recovery by using the electrocoagulation process. Therefore, in the available research, RSM is carried out to optimization of effective factors on Cu metal recovery and produced sludge volume in thickener overflow by using D-optimal experimental design.

The purpose of the present study is copper recovery from overflow of copper concentration plant by electrocoagulation method and So far no studies have been conducted on copper recovery using electrocoagulation in mineral processing plants. Most studies have performed to copper removal from industrial effluents.

Moneer et al. (2015) have studied on removal of copper from simulated wastewater by electrocoagulation/floatation technique. Results indicated that the initial concentration would be less than the minimum time required to remove 100% of copper from the wastewater, and optimal conditions were considered in each experiment [31]. Ferreira et al. (2013) are researched on the removal of copper, zinc and nickel present in natural water containing Ca^{2+} and HCO_3^- ions by electrocoagulation. In this study, only 95% of copper was removed in 60 minutes [25]. Kamaraj et al. (2012) are studied the effect of direct current (DC) and alternating current (AC) on the removal of copper from the water by the electrocoagulation process. The results showed that when the pH value was 7 and the current density was 0.025 mA / cm^2, the optimal copper removal in the DC was 97.8% and in the AC was 97.2% [17]. Akbal and Kamsi (2011) are conducted a study on Copper, chromium and nickel removal from metal plating wastewater by electrocoagulation. The results showed that combination of iron and aluminum electrodes, removal of 100% copper for 20 minutes at a current density of 10 mA / cm^2 and pH = 3 was obtained [20].

In according to the mentioned studies, the electrocoagulation process has been considered as an effective method for the separation of copper in aqueous solutions, so that in all these studies the copper removal efficiency is above 95% which it can be concluded that this system is an applicable method in industrials and especially in mineral processing plants for the recovery of copper.

2. Materials and methods

2.1. Experimental procedure

Water samples were collected from the thickener overflow (included fine concentrate with 30% grade) that to discharge in electrocoagulation reactor. Electrocoagulation process have created two products that contain Cu concentrate and water recycling. Schematic planning of electrocoagulation system is underscored in Figure 1. Samples were conserved in polyethylene vessel and cooled down to 4ºC. These samples were tested with standard analytical methods.

Electrocoagulation system was implemented in an electrochemical reactor and batch model with capacity of 6000 mL (measuring 30 × 20 × 10 cm). Electrocoagulation reactor has an effective volume of 5 L which is created from Plexiglas. Numbers of 6 electrodes from Iron and aluminum material with a 20*2 cm^2 dimension were applied as electrodes. The area of influence electrodes surface was noted 90 cm^2 and the distance between electrodes [cathode and anode] was considered 2 cm. The electrodes need to wash with HCl (15%) followed by distilled water before to each test. The cell was cleared by washing twice with distilled water. The water sample from thickener overflow was added to the electrocoagulation cell after providing a direct current power supply and put the electrodes in their place. However, samples were regulated to initial pH by hydrochloric acid or sodium hydroxide before each experiments run.

In electrocoagulation process, produced coagulations transfer to surface and sludge produce in this area and then produced sludge by a 0.45 mm filter is vacuum filtered. it was then dried at 105ºC for 24 h and also the Cu grade percentage in sludge was characterized by UV-Visible Spectrophotometer where 0.2 g/L Cu (II) solution in the electrocoagulation cell was taken at different conditions. Therefore, the effect of different variables on the % Cu (II) grade and produced sludge volume were investigated.

2.2. Analytical methods

Copper grade is the concentration of copper in the sludge. To calculate the grade of copper for the samples, applying the follow equation:

$$\text{Cu grade} = \frac{\text{amount of copper metal}}{\text{amount of Sludge containing copper}} * 100 \qquad (1)$$

Whereas, produced sludge volume is taken from following equation:

$$\text{sludge volume} = S * H \qquad (2)$$

Where, S is available sludge surface (cm^2) and H is high sludge of produced process (cm)

2.3. Experimental design

In according to literature reform and opening up, comprehensive investigations don't perform on the electrochemical system for the Cu recovery from mine industrials (mineral processing) by response surface methodology (RSM).

Thus, the purpose of investigation is to study the effects of process parameters on Cu grade using electrocoagulation with different electrodes. Operation parameters can extremely effect on the electrocoagulation performance, namely initial pH, Current density, and electrolysis time. Hence, in this research, these parameters are investigated and optimized for Cu recovery and produced sludge volume with response surface methodology (RSM). RSM is a generally implemented statistical instrumentation where major variables effect some response(s), and with regard to available situation, optimization process is desirable. The main advantage of RSM is that, in opposition to one factor-at-a-time survey, experiments is demanded with fewer number and the interaction between variable could be studied [43, 44].

In this research, the influence of procedure variables was utilized to study experimental design of D-optimal with four agents [three numerical and one categorical variables] at three levels. Studied variables were included

electrolysis time (X_1), initial pH (X_2), current density (X_3) and electrodes type (X_4) in order to investigate the Cu grade (Y_1) and sludge volume (Y_2) from thickener overflow using electrocoagulation technique (Table 1).
The selection of independent variables deduced in experiments which are expressed as given below:

$$X_i = \frac{x_i - x_0}{\Delta x_i} \qquad i = 1,2,\ldots,k \tag{3}$$

Where X_i is considering as the dimension minor-encoded quantity of an independent changing; x_i is containing the actual quantity of an independent changing; x_0 is the actual quantity at the center point of an independent changing; and Δx_i includes the stage conservation of the actual quantity of the changing $i.$, On this basis, the information resulted from the equation were exploited to obtain a prediction model. Experimental later-order multinomial pattern is exhibited in [Eq. [4]]:

$$\gamma = \beta_0 + \sum_{j=1}^{k} \beta_j x_j + \sum_{j=1}^{k} \beta_{jj} x_j^2 + \sum \sum_{i<j=2}^{k} \beta_{ij} x_i x_j \tag{4}$$

where γ is the response from this condition; variables are x_i & x_j [i & j varied from 1 to k]; constant phrase isβ_0 ; linear coefficient is β_j, interaction factor is β_{ij}, and quadratic factor is β_{jj}; number of independent parameters is k that in available condition k equal 4 [45,46].

Table 1. Range of independent variables and their levels.

Variable (unite)	Numerical variable		
	Level		
	-1	0	1
X_1, Electrolysis time (min)	5	10	15
X_2, pH	4	7	10
X_3, Current density (A/m^2)	41.6	104.1	166.6
	Categorical variable		
	Level		
	1	2	3
X_4, Electrode type	Fe	Fe -Al	Al

3. Results and discussion

3.1. D-optimal design

D-optimal plans are presented as one shape of project using a computer for problem-solving of operations pattern..These kinds of assisted designs are exclusively beneficial when classical designs do not handle. In opposite standard classical plans, namely factorials and fractional factorials, D-optimal design matrices are generally non-orthogonal and estimations have important effect on correlation.
That is reason using D-optimal designs commonly stand in two classifications: the standard factorial design implicates multimadia runs with quantity of resources or permissible period for the examination or the plan span is compelled; i.e. the process span includs factor settings that are not practical or are inconceivable to run [47].

3.1.1. Evaluation of experimental results

In according to the produced design, 20 experiments were executed and the details of the experimental conditions are presented in table 2. The equations of model regression [1] and [2] were provided using Design of Expert [7.0.0] and are determined as follows:

$$\text{Cu grade, [\%]}\ Y_1 = 19.74 - 6.60X_1 - 1.02X_2 + 1.13X_3 + 2.85X_4 - 1.72X_1X_2 + 2.12X_1X_3 - 12.34X_1X_4 - 0.43X_2X_3 - 10.45X_2X_4 + 8.89X_3X_4 - 0.22X_1^2 - 2.45X_2^2 + 2.01X_4^2 + 9.76X_1X_2X_4 - 11.74X_1X_3X_4 - 15.65X_2X_3X_4 \tag{5}$$

$$\text{Sludge Volume, [cm}^3\text{]}\ Y_2 = 343.34 + 207.20X_1 - 63.77X_2 + 27.83X_3 - 59.20X_4 - 22.35X_1X_2 - 6.56X_1X_3 - 10.96X_1X_4 + 35.77X_2X_3 + 29.56X_2X_4 - 4.16X_3X_4 + 17.14X_1^2 + 35.08X_2^2 + 161.66X_3^2 - 131.24X_4^2 \tag{6}$$

Table 2. Experimental design matrix and response based on the experimental runs on Cu grade (%) and sludge volume by D-optimal design.

Run no.	X_1: (min)	X_2:	X_3: (A/m^2)	X_4:	Cu grade (%)	Sludge Volume (cm^3)
1	10	7	104.1	Fe	24.25	180
2	5	7	104.1	Fe-Al	27.03	180
3	15	4	166.6	Fe	23.26	600
4	15	10	41.6	Fe	15.34	450
5	15	4	166.6	Al	15.15	810
6	15	7	104.1	Fe-Al	11.8	540
7	10	10	41.6	Al	18.41	300
8	10	7	104.1	Fe-Al	20.29	300
9	5	10	166.6	Al	24.75	270
10	5	4	41.6	Fe	24.75	180
11	5	4	166.6	Fe-Al	22.37	390
12	5	4	41.6	Al	26.43	330
13	5	10	166.6	Fe	24.75	210
14	15	4	41.6	Fe-Al	10	870
15	15	10	166.6	Fe-Al	10.5	750
16	15	10	104.1	Al	19.9	420
17	5	10	41.6	Fe-Al	25.25	240
18	10	10	104.1	Fe-Al	17.18	330
19	15	7	41.6	Al	18.31	660
20	10	4	104.1	Fe-Al	17.2	450

The resulted D-optimal data was assessed by two various examinations such as the sequential model sum of squares and model summary statistics in order to achieved influence regression models from between different models, namely linear, interactive, quadratic and cubic. The conclusions of the Cu grade percent and sludge volume are demonstrated in tables 3(a) and (b). With regard to results, it is apperceived that, linear model displays in cooperation of other models (quadratic and interactive (2FI); increasing R^2, regulated-R^2, predicted-R^2, F-value and low p-value. The cubic model was established to be aliased and could not be utilized for subsequent modeling from experimental data. Aliased model can be means that not sufficient experiments have been run to independently evaluation total the terms for this model. Whatever, there are more less independent points in the scheme than there are terms in the model, some parameters cannot be calculated independently. Hence, the linear model is selected to demonstrate the influence of process variables on the electrocoagulation process to Cu processing.

Meanwhile, the model summary statistics represented that after preventing the cubic model which was aliased, the linear model was established to have the maximum "adjusted R^2" and predicted R^2" values. Simultaneously, the analysis was performed for sludge volume and the results are showed in table 3(a). Therefore, the quadratic model was selected to express the influence of process variables on the resulted sludge volume from the electrocoagulation process.

Table 3. Sequential model sum of squares and model summary statistics for Cu grade, (%).

Source	Sum of Squares	Degree of freedom	Mean Square	F Value	Prob > F	
Mean vs Total	7877.27	1	7877.27			
Linear vs Mean	339.27	4	84.82	6.22	0.0037	Suggested
2FI vs Linear	45.25	6	7.54	0.43	0.8444	
Quadratic vs 2FI	122.43	4	30.61	4.15	0.0754	
Cubic vs Quadratic	36.9	5	7.38			Aliased
Residual	0	0				
Total	8421.12	20	421.06			

Model summary statistics

Source	Std. Dev.	R-Squared	Adjusted R-Squared	Predicted R-Squared	PRESS	
Linear	3.69	0.6238	0.5235	0.3075	376.61	Suggested
2FI	4.21	0.707	0.3815	-3.7505	2583.52	
Quadratic	2.72	0.9321	0.7422	-0.8925	1029.25	
Cubic					+	Aliased

+ case with leverage of 1.00 : PRESS statistic not defined

Table 4. Sequential model sum of squares and model summary statistics for sludge volume, (Cm^3).

Source	Squares	Degree of freedom	Square	Value	Prob > F	
Mean vs Total	3.58E+006	1	3.58E+06			
Linear vs Mean	6.85E+05	4	1.71E+05	12.71	0.0001	
2FI vs Linear	50555.47	6	8425.91	0.5	0.7941	
Quadratic vs 2FI	1.46E+05	4	36568.72	34.29	0.0008	Suggested
Cubic vs Quadratic	5331.92	5	1066.38			Aliased
Residual	0	0				
Total	4.47E+06	20	2.23E+05			

Model summary statistics

Source	Std. Dev.	R-Squared	Adjusted R-Squared	Predicted R-Squared	PRESS	
Linear	116.09	0.7721	0.7113	0.6131	343239.3	
2FI	129.78	0.8291	0.6392	-0.5499	1375147	
Quadratic	32.65	0.9939	0.9771	0.7913	185117	Suggested
Cubic					+	Aliased

+ case with leverage of 1.00 : PRESS statistic not defined

3.1.2. ANOVA results

The importance and competency of the model were evaluated by analysis of variance (ANOVA). ANOVA compares variation sources with Fisher dissemination (F-test) to validate the reliability of the regression model. High F-value of 18.75 and 59.07 for copper grade and sludge volume, respectively that pointed the admissibility of the change about its mean values. It obviously illustrated the predictability of the regression model at the 95% confidence interval. T-Test was used to study the important of each coefficient of the developed regression model (presented in Equation (5, 6)) and corresponding p-values for Cu grade and sludge volume was found 0.001 and 0.0168 respectively. The probability of error value (p-value) is utilized to detect whether the community between the response and each term in the model is statistically significant. The p-value is identified as the smallest level of significance leading to rejection of the null hypothesis and the interaction influences are statistically significant when $p < .05$ [48, 49].

In this condition, Parameters including p-values> .05 can be omitted using the step by step elimination method. Low values of 6.75 for Cu grade and also 7.72 for sludge volume, in coefficient of variation (C.V.) determined a higher degree of accuracy and validity of the experiments.

The adequate accuracy value is a measure of the "signal to noise ratio" for the responses. A ratio which is more than 4 was inspected as adequate model discernment [50].

In this research, the signal-to-noise ratio for the Cu grade and sludge volume was acquired to be 13.68 and 25.182, respectively, which illustrated there was adequate signal. Hence, the linear and quadratic models could be applied to navigate the design term.

Results demonstrate the analysis of variance (ANOVA) of regression variables depending to the estimated response surface linear model for Cu grade and quadratic model for sludge volume, respectively. On consideration of results, they are comprehended that the fisher's F-tests with a low probability value [for Cu grade: p-value = < 0.0168 and for sludge volume: p-value = < 0.0001] indicates that the regression models is statistically significant. It is clear that the model design are significant when values of prob>F are less than 0.05. Therefore, in this study, the p-values were lower than 0.05. The other standard utilized for assessing the model was lack-of-fit [LOF] test, which is implemented for evaluating the residual with pure error attended from the frequented design points at the central level of variables. As it can see, if the F-value of LOF is lower than F-Table or the depended p-value is more than 0.05 the regression will adequately be significant. In this condition the significance level is considering 95% [51, 42].

It illustrates the ratio of the whole variance of the dependent variable clarified using the regression pattern. High coefficient determination R^2 proves that the quadratic model expresses total of the variations of the dependent variable. In this research, the R^2 value of the models determined that 99.01% and 99.40% of the total variability could be depicted by the models for the Cu grade and sludge volume, respectively. The values of the adjusted designation coefficient (adjusted R^2 = 0.9373 and 0.9772) demonstrated the importance of both Cu grade and sludge volume.

Table 5. ANOVA analysis for the linear model developed for Cu grade from electrocoagulation process.

Source	Sum of Squares	Degree of freedom	Mean Square	F Value	p-Value Prob > F	
Model	538.46	16.00	33.65	18.75	0.0168	significant
X_1	150.56	1.00	150.56	83.88	0.0028	
X_2	3.62	1.00	3.62	2.01	0.2508	
X_3	3.26	1.00	3.26	1.82	0.2706	
X_4	6.24	1.00	6.24	3.48	0.1591	
X_1X_2	7.60	1.00	7.60	4.23	0.1318	
X_1X_3	10.70	1.00	10.69	5.95	0.0925	
X_1X_4	6.34	1.00	6.34	3.53	0.1567	
X_2X_3	0.44	1.00	0.44	0.25	0.6538	
X_2X_4	4.55	1.00	4.55	2.53	0.2096	
X_3X_4	3.21	1.00	3.21	1.79	0.2733	
X_1^2	0.13	1.00	0.13	0.07	0.8083	
X_2^2	15.39	1.00	15.39	8.57	0.0611	
X_3^2	2.81	1.00	2.81	1.57	0.2994	
$zX_1X_2X_4$	2.61	1.00	2.61	1.46	0.3141	
$X_1 X_3X_4$	3.86	1.00	3.86	2.15	0.2390	
$X_2X_3X_4$	6.86	1.00	6.86	3.82	0.1456	
Residual	5.39	3.00	1.80			
Cor Total	543.85	19.00				
Std. Dev.	1.34	R^2				0.9901
Mean	19.85	Adj. R^2				0.9373
*C.V%	6.75	Adeq. precision				13.680
*PRESS	235.81					

*C.V. %: Coefficient of variation, PRESS : Predicted residual sum of squares

Table 6. ANOVA analysis for the linear model developed for sludge volume produced from electrocoagulation process.

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	881900.00	14.00	62992.01	59.07	0.0001	significant
X_1	580700.00	1.00	580700.00	544.58	<0.0001	
X_2	55002.74	1.00	55002.74	51.58	0.0008	
X_3	8538.45	1.00	8538.45	8.01	0.0367	
X_4	36401.53	1.00	36401.53	34.14	0.0021	
X_1X_2	5180.98	1.00	5180.98	4.86	0.0787	
X_1X_3	456.47	1.00	456.47	0.43	0.5418	
X_1X_4	1004.33	1.00	1004.33	0.94	0.3764	
X_2X_3	13586.19	1.00	13586.19	12.74	0.0161	
X_2X_4	7312.62	1.00	7312.62	6.86	0.0472	
X_3X_4	138.90	1.00	138.90	0.13	0.7329	
X_1^2	639.84	1.00	639.84	0.60	0.4736	
X_2^2	2679.07	1.00	2679.07	2.51	0.1738	
X_3^2	43203.17	1.00	43203.17	40.51	0.0014	
X_4^2	68263.73	1.00	68263.73	64.01	0.0005	
Residual	5331.92	5.00	1066.38			
Cor Total	887200.00	19.00				
Std. Dev.	32.66	R^2				0.9940
Mean	423.00	Adj. R^2				0.9772
*C.V%	7.72	Adeq. precision				25.182
*PRESS	185100.00					

*C.V. %: Coefficient of variation, PRESS: Predicted residual sum of squares

3.1.3. Experimental versus predicted

Competency survey of the suggested model is a significant section of the analytical process. Respectable competency confirms that the approaching model produces a competency approximation to prevent weak or incorrect out comes. The evaluation between the experimental and predicted value from the model is presented in Figure 2(a) and (b). It was proved that the model predictions coincided the experimental values and the data points set nearby to the diagonal line. The analysis displayed that these second order multinomial equations could desirable predict for the Cu grade and sludge volume by electrocoagulation process.

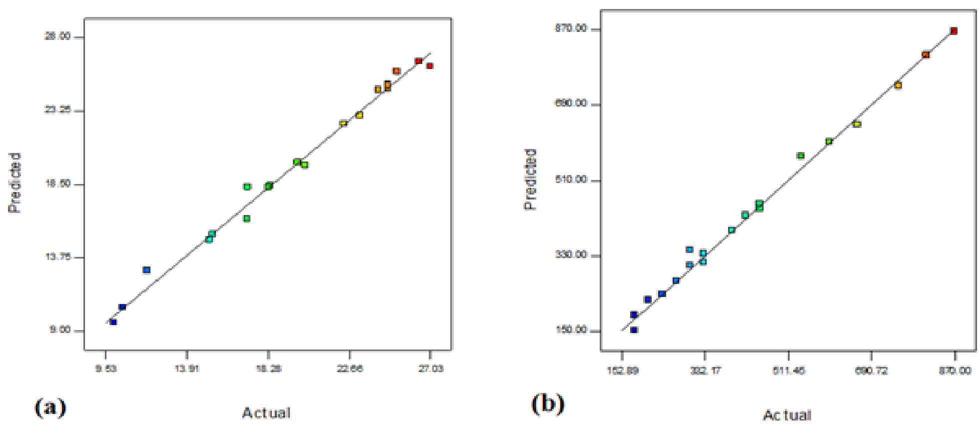

Figure 2. The actual and predicted plot for (a) Cu grade and (b) sludge volume.

3.1.4. Normal probability

The residuals determined how the model approved the hypotheses of ANOVA, whilst the standardized residuals evaluated with the standard deviations for separating the observed and predicted values. The data were also assessed to determine the normality of the residuals that obtained results are underscored in Figure 3(a) and (b). The Figure 3(a) and (b) explains the normal% probability plots of the standardized residuals for (a) Cu grade percentage and (b) sludge volume using electrocoagulation process. A normal probability plot examines whether the residuals pursue a distribution in normal, in which case the points will create a straight line. Some dispersion are approached even with normalized data [52, 53]. It can hence be comprehended from Figure 3(a) and (b) that the data is distributed in normally.

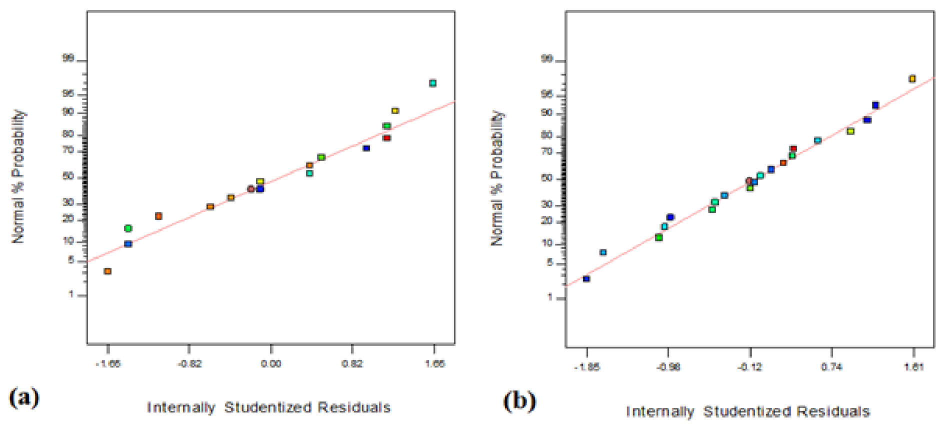

Figure 3. Plot for relationship between normal% probability and external studentized residuals for (a) % Cu grade and (b) sludge volume.

3.2. Effects of process parameters

Major electrochemical processes are considering pH, current density, and electrolysis time which are most significant parameters for adjusting the reaction rate [54, 49]. Thus, the influences of three main parameters on the Cu grade and sludge volume were studied using the RSM. The main advantage of using RSM is influences of the reactions between parameters which could be underscored.

3.2.1. Initial pH

pH is a significant parameter for optimization of electrocoagulation process because it influences on the solution conductivity, zeta potential and dissolution of electrode. However, it is too problem to obtain a correct relationship between the solution pH of the solution and the efficiency of electrocoagulation process since pH of the processed water varies in during electrocoagulation process, thus it is normally originated to the initial solution pH [55-58].

The obtained results of the effects of pH on Cu grade percentage and sludge volume by electrocoagulation process are illustrated as the 3D surface graph in Figure 4(a and b) and Figure 5(a and b). The pH of the experiments was changed in the range of 4–10. The results in

Figure 4 (a) proved that the Cu grade efficiency increased at pH: 4-7 with reducing electrolysis time and highest efficiency was obtained 96% (with grade of % 27.03) at pH: 7 by Fe as anode and Al as cathode electrodes, but in Figure 4 [b], Cu grade efficiency increased at pH: 10 which it trend to pH 4 with increased current density.

Figure 5 (a) displays the effects of pH on the sludge volume which with increasing initial pH and current density, the produced sludge volume also increases. It can be underscored from Figure 5 (b) the sludge volume slightly increased when the effluent pH was increased from pH: 10 to pH: 4 but this increasing is more pronounced in the current density 41 and 166 A/m^2. This could be due to decrease electrolyte conductivity when the effluent pH varied from acidic to neutral situation and vice versa [53].

This can be expressed as reactions between iron anode and aluminum cathode by electrocoagulation process according to the following equations:

For the, anodic reaction,

$$Fe_{(s)} \rightarrow Fe^{2+}{}_{(aq)} + 2e^- \quad (7)$$

Cathodic reaction,

$$2H_2O_{[l]} + 2e^- \rightarrow H_{2(g)} + 2OH^-{}_{[aq]} \quad (8)$$

Solution reaction,

$$Fe^{2+}_{(aq)} \rightarrow 2OH^-{}_{[aq]} + Fe(OH)_{2[S]} \quad (9)$$

Overall reaction,

$$Fe^{2+}_{(s)} \rightarrow \; 2H_2O_l \rightarrow Fe(OH)_{2(S)} + H_{2(g)} \quad (10)$$

Iron could exists as Fe $^{2+}$ or Fe^{3+} cations, which this iron also can hydrolyzed as insoluble iron that depending to the solution pH and the potential of cell (Eq. (7) & (10)). The anodic reactions create some acidic situation in around of anode, which is disagreed, using the cathode in around of alkaline environment owing to hydrogen evolvement and formation of OH^- ions (Eq. (10)). As a comprehensive approach, when the primary pH of the solution is extremely acidic (pH < 3) or extremely alkaline (pH > 11) there is no significant variation in the primary pH. However, when the primary pH is acidic, pH should increase by the electrocoagulation process and when the primary pH is alkaline, pH should reduce in during the electrocoagulation process. The consequence defined that iron electrolysis cause to the production of Fe^{2+} that then expose oxidation in presence of dissolved oxygen and appropriate pH to produce Fe^{3+} that in finally is formed insoluble $\mathrm{Fe[OH]_{3[S]}/FeOOH_{[S]}}$. In electrocoagulation process, the formation of OH^- ions at the cathode provisional raises the pH before it expended by the Fe^{2+} formed at the anode, which accelerate to the reaction rate of Fe^{2+} oxidation to Fe^{3+}. pH then declines as OH^- ions are expended at the anode. It was also detected that at proportion down pH (4.5-6.5) the rate of Fe^{2+} oxidation and hydrolysis is gradual which outcome an enhance in the solution pH and production of a combination of soluble ferrous ions and insoluble $\mathrm{Fe[OH]_{3[S]}/FeOOH_{[S]}}$ [59]. Whatever the pH of the solution lead to alkaline position (7.5-10) sedimentation ratio and oxidation of Fe^{2+} to Fe^{3+} is slowly happen .

Produced amorphous $Al[OH]_{3[s]}$ flocs at pH limited area 6.5–7.8 have an extended particular surface in condition least solubility which have capability absorb some soluble organic compounds [60, 42]. The maximum Cu grade percentage is attained in relatively neutral medium.

It was derived that in normal situation, most hydroxyl radicals were created in acidic and alkaline mediums of the hybrid electrocoagulation process [61]. Same resultant on the influence of pH on the pollutants removal from industrial areas have been perceived in numerous investigations [62, 53].

Obtained results are in admirable consent with the discovery of other investigations that the pH could be more than 5 if ferrous ions have ability only oxidized to ferric ions [55, 57]. In much basic pH, unsuitable $Fe(OH)_4{}^-$ forms which is considering an incapable coagulation material that cause to destroy electrocoagulation proceeds [55]. It is deduced that pH range of the optimum in operating condition is 6-9 for iron electrocoagulation and operating condition is suitable to assure perfect oxidation of ferrous ions at an initial pH of 6.5-7.5 which are extremely soluble, weak coagulants with very low adsorption ratio.

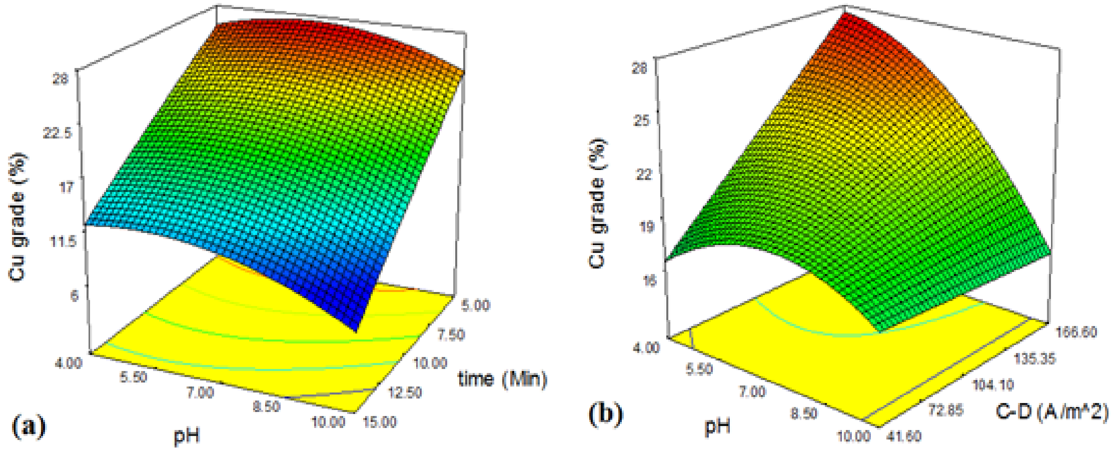

Figure 4. Effect of a) initial pH and electrolysis time, (electrode type: Fe-Al; current density: 104.10) and b) initial pH and current density (electrode type: Fe-Al; electrolysis time: 10 min) on Cu grade (%), 3D surface plots.

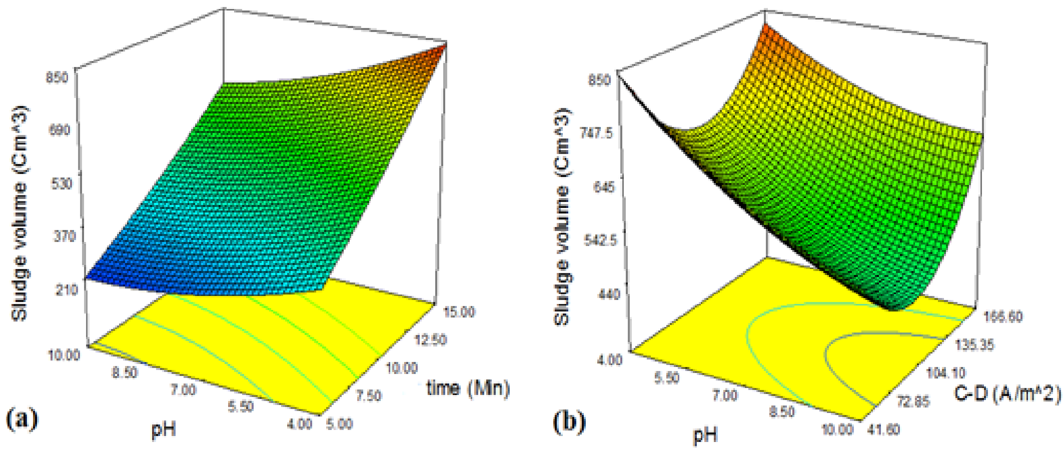

Figure 5. Effect of a) initial pH and electrolysis time (electrode type: Fe-Al; current density: 104.10), and b) initial pH and current density (electrode type: Fe-Al; electrolysis time: 10 min) on sludge volume (Cm3); 3D surface plots.

3.2.2. Current density

One of the critical parameters for improving electrocoagulation process is current density. It shows the amount of metal ions desorbed from the electrodes and is an important value for current density which if exceeded the quality of processed water does not display significant improvement. The choice of an optimum amount for current density is also influenced by other parameters such as pH and electrolysis time [63, 58]. Various researchers investigated the influence of current density on the operation system [64-66, 54, 55].

Figures 6(a,b) and 7 (a,b) represents 3D plots and 2D contours to studied the influence of current density and electrolysis time on cu grade percentage and produced sludge volume. The results explain with increasing the current density, the Cu grade percent and sludge volume increases. This is ascribed to an increment in the value of Fe (III) by anode dissolution, which has an intense dependence toward inorganic matter and provides desirable adsorption sites for Cu. In addition; more production of hydrogen permitted by upper current aids the flotation of the flocculation matter [46].

Also, the dissolution rate of iron increased with the increase in current density and thus a fixed amount of Cu reacted at 166.6 A/m^2 current density and 5-7 minute. However with increasing electrolysis time, Cu grade percentage increasing and Cu grade reduced. That is why; the whole of Cu grade is resulted in initial time of 5 minutes reaction. With increasing electrolysis time, organic matter and anode expended rising in sludge which cause to reduce of Cu grade percentage in sludge.

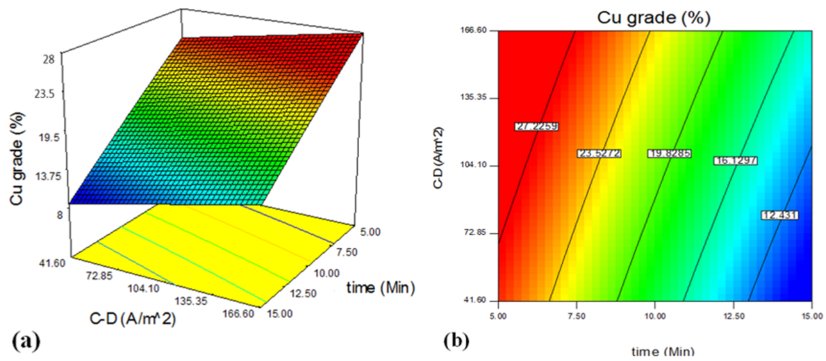

Figure 6. Effect of current density and electrolysis time on Cu grade (%),a) 3D plots and b) 2D contours.

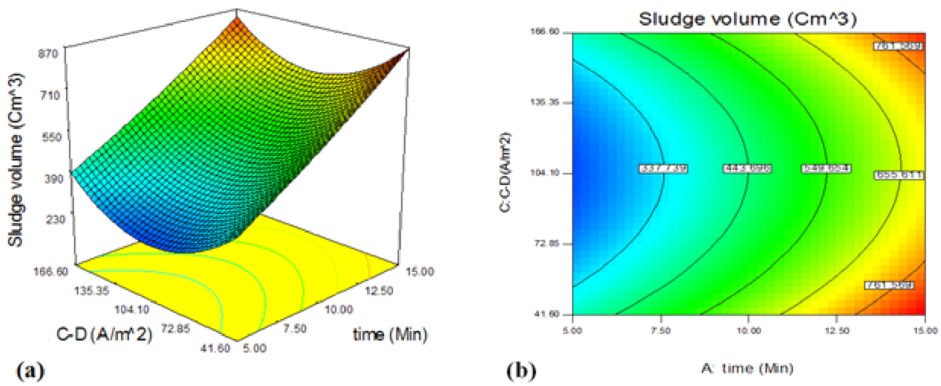

Figure 7. Effect of current density and electrolysis time on sludge volume, a) 3D plots and b) 2D contours.

3.2.3. Electrolysis time

Electrolysis time is a major parameter for cu grade percent and produced sludge volume electrocoagulation process [59, 16]. The experiments were executed by varying electrolysis time (X_1) from 5 to 15 min with: pH: 7 (x_2), current density =104.10 A/m^2 (x_3), Electrode type = Fe-Al (x_4). The results are depicted in Figure 8(a,b), which illustrates that the optimum time for cu grade percent is 5 min and produced sludge volume is obtained at 15 min.

The result demonstrates that cu grade percentage reduced with increasing electrolysis time because of the whole of Cu grade is derived in initial time of 5 minutes reaction. With increasing electrolysis time, organic matter and anode expended rising in sludge which cause to reduce of Cu grade percentage in sludge. However, increasing electrolysis time cause to increasing produced sludge volume. This is due to iron electrolysis cause to convert Fe^{2+} to Fe^{3+} and it produces metallic ions which are appropriate coagulants. At the cathode, OH− ions are formed during water electrolysis, and they can react with the metallic ions to produce sludge [67-69].

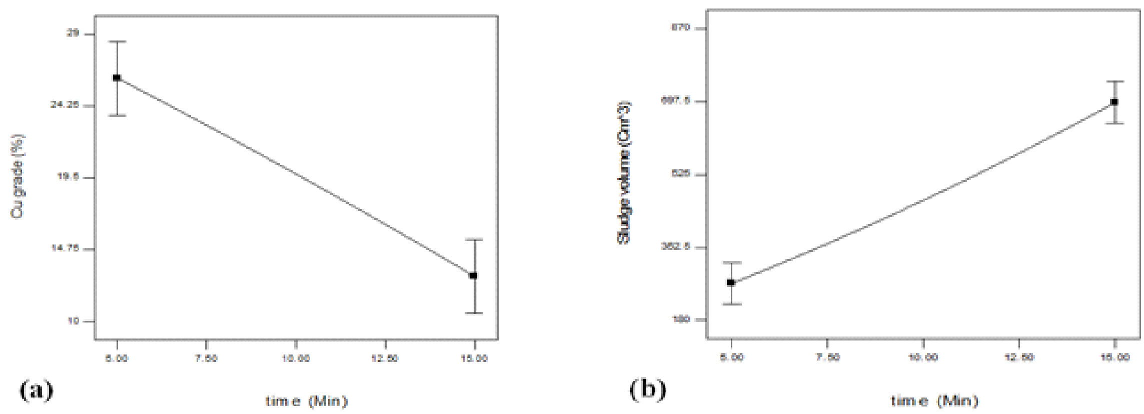

Figure 8. Effect of time a) on Cu grade (%) and b) on sludge volume (electrode type: Fe-Al; current density: 104.10; initial pH: 7), One factor plot

3.3. Optimization and validation

One of the principal objectives of this study is to obtain the optimal conditions for the maximum of Cu grade and sludge volume from thickener overflow using electrocoagulation process. The obtained yields in order to optimization of process were utilized the regression equation of RSM based on the D-optimal. In the optimization process of electrolysis time (X_1), pH (X_2), current density (X_3), electrode type (X_4) were choiced as within range and the responses such as Cu grade percentage(Y_1) was maximized and produced sludge volume (Y_2) was maximized, too.

In according table 4, the optimization of experimental conditions, The Cu grade percentage and sludge volume was attained to be 27.03% and 870 cm^3, respectively which are authenticated under the optimal conditions by creating additional experiments. A mean value of 19.84% for Cu grade and 423 cm^3 for sludge volume were taken from the experimental, which is in nearby compromise with the predicted values attained. The suitable correlation between these actual and predicted results indicate that the reliability of D-optimal design is containing desirability function method and it could be effectively utilized to optimize electrocoagulation process parameters for any type of Cu particles in mineral processing plant.

4. Conclusions

In this research, D-optimal was executed to investigate and optimize the process variables such as electrolysis time, initial pH, current density and electrodes type on the initial Cu grade percentage (28%) and produced sludge volume from thickener overflow using electrocoagulation process in mineral processing plant.

The results illustrated that in most electrochemical processes; electrolysis time, pH, and current density are the majority parameters for adjusting the reaction ratio. High F-value of 18.75 and 59.07 are created for copper grade and sludge volume, respectively that pointed the admissibility of the change about its mean values. It obviously illustrated the predictability of the regression model at the 95% confidence interval. Low values of 6.75 for Cu grade and also 7.72 for sludge volume, in coefficient of variation (C.V.) determined a higher degree of accuracy and validity of the experiments. The linear and quadratic models could be applied to navigate the design term. It was proved that the model predictions coincided the experimental values and the data points set nearby to the diagonal line. A normal probability plot examines whether the residuals pursue a distribution in normal, in which case the points will create a straight line.

The pH of the experiments was changed in the range of 4–10. The results proved that the Cu grade efficiency increased at pH: 4-7 with reducing electrolysis time and highest efficiency was obtained 96% (with grade of % 27.03) at pH: 7 by Fe as anode and Al as cathode electrodes, but Cu grade efficiency increased at pH: 10 which it trend to pH: 4 with increased current density. The sludge volume slightly increased when the initial pH was increased from 10 to 4 but this increasing is more pronounced in the current density 41 and 166 A/m^2.

The results are explained with increasing the current density, the Cu grade percent and sludge volume increases. Also, the optimum time for cu grade percent is 5 min and produced sludge volume is obtained at 15 min. A mean value of 19.84% for Cu grade and 423 cm^3 for sludge volume were taken from the experimental, which is in nearby compromise with the predicted values attained.

The actual optimal values of electrolysis time, pH, current density and electrode type resulting in 27.22% Cu grade were 6.5 min, 6.7, 50.2 A/m^2 and Fe-Al, respectively. Similarly, for the optimization of experimental conditions for produced sludge volume (861 cm^3) was found electrolysis time: 15 min; initial pH: 4.1; current density: 48.7; electrode type: Fe-Al.

The results showed the technical feasibility of electrocoagulation known as a practicable and reliable technique for Cu recovery from mine industrials especially in mineral processing plant. This research establishes the stand of more research on other extend span of variables and comprehensive review which are essential to inquire these results for application of mining industry that lead to optimization in large-scale of the electrocoagulation process.

Table 7. Comparison of verification and predicted values of Cu grade and produced sludge volume by electrocoagulation at maximum optimum condition.

Response	optimum condition					
	X_1	X_2	X_3	X_4	Experimental	Predicted
%Cu grade	6.21	6.78	50.28	Fe-Al	27.22	28.05
Sludge volume	15	4.01	48.7	Fe-Al	861	982.78

Notes: Where X_1 = electrolysis time (min), X_2 = initial pH, X_3 = current density (A/m^2), X_4= electrode type.

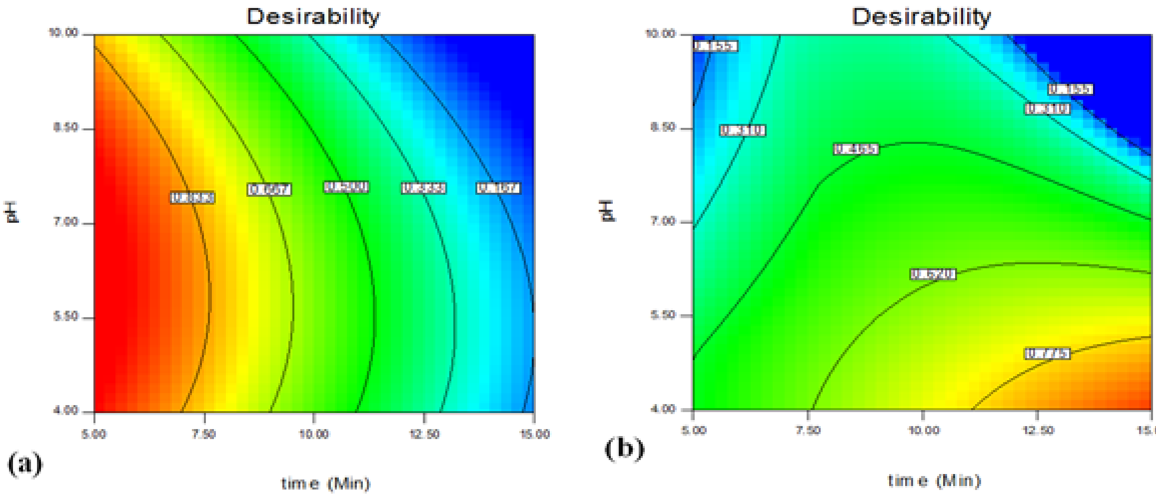

Figure 9. Desirability to obtain (a) Maximum Cu grade and (b) Maximum sludge volume (electrode type: Fe-Al; current density: 104.10), 2D contours.

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بازیابی مس از سرریز تیکنر با استفاده از الکتروکواگولاسیون/ شناورسازی: بهینه‌سازی با استفاده از سطح پاسخ، مدل‌سازی و مطالعه لجن

علی حسن زاده سبلوئی[1] و سید مرتضی موسوی راد[2*]

۱- بخش مهندسی معدن، مجتمع آموزش عالی زرند، ایران

۲- گروه پژوهشی محیط‌زیست، پژوهشگاه علوم و تکنولوژی پیشرفته و علوم محیطی، دانشگاه تحصیلات تکمیلی صنعتی و فناوری پیشرفته، کرمان، ایران

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* نویسنده مسئول مکاتبات: s.m.moosavirad@kgut.ac.ir

چکیده:

فرآیند الکتروکواگولاسیون/ شناورسازی یک روش جدید در صنعت معدن است که برای بازگشت فلز مس به چرخه تولید می‌تواند مورد استفاده قرار گیرد. این روش باعث بهبود بازیافت مس و کاهش پساب می‌شود. در این پژوهش، از روش سطح پاسخ برای بهینه‌سازی عوامل مؤثر در بازیافت فلز مس و حجم لجن تولید شده از سرریز تیکنر استفاده شده است. برای این منظور، از طرح آزمایشی D-optimal استفاده شد. تأثیر چهار پارامتر مستقل از جمله زمان الکترولیز، pH اولیه، چگالی جریان و نوع الکترود برای بررسی درصد عیار مس (۲۸٪) و حجم لجن تولید شده از سرریز تیکنر مورد بررسی قرار گرفت. همه این پارامترها اثرات مهمی در بازیافت فلز مس و میزان لجن تولید شده دارند. از مدل‌های خطی و درجه دوم به ترتیب برای مقادیر مس و حجم لجن استفاده شد. اهمیت متغیرهای مستقل و تعامل بین آن‌ها توسط ANOVA ارزیابی شد. شرایط عملیاتی بهینه با عیار مس ۲۷/۲۲ درصد در نظر گرفته شد: زمان الکترولیز: ۶/۵ دقیقه، pH اولیه: ۶/۷، چگالی جریان: ۵۰/۲ آمپر در مترمربع و نوع الکترود: آهن-آلومینیوم. به طور مشابه، برای حجم لجن تولید شده $۸۶۱cm^3$، شرایط زیر یافت شد: زمان الکترولیز: ۱۵ دقیقه، pH اولیه: ۴/۱، چگالی جریان: ۴۸/۷ و نوع الکترود: آهن-آلومینیوم. نتایج با تأکید بر یک دیدگاه عملی از الکترووا‌ژولاسیون، به عنوان یک روش قابل قبول برای بازیابی مس از صنایع معدنی، به ویژه در کارخانه‌های فرآوری مواد معدنی شناخته شده است.

کلمات کلیدی: بازیابی مس، الکتروکوآگولاسیون/ شناورسازی، صنایع معدنی، طرح آزمایشی D-optimal، بهینه‌سازی.