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Electrochemical removal of copper from simulated wastewater using a rotating tubular packed bed of woven screens electrode

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ABSTRACT

Copper removal from simulated wastewater was investigated by using a rotating tubular packed bed of woven screens electrode as a cathode in a new design of the electrochemical reactor. The effects of electrolysis operating parameters like current (0.5–2.5 A), rotation speed (150–750 rpm), and initial copper concentration (100–500ppm) were investigated. Optimization of process parameters was carried out by adopting response surface methodology (RSM) combined with Box–Behnken Design (BBD), where copper removal efficiency was selected as a response function. The results indicated that the current has the main effect on the copper removal efficiency followed by rotation speed and concentration. The results of regression analysis revealed that the experimental data could be fitted to a second-order polynomial model with a value of determination coefficient (R^2) equal to 0.9894 and Fisher test at a value of 51.57. The optimum conditions of the process parameters based on RSM method were an initial copper concentration of 205 ppm, current of 2.5A, and rotation speed of 750 rpm utilizing cathode composed of screens with mesh no. 30 where a final copper concentration less than 2 ppm was obtained after 30 min.

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1. Introduction

Copper considers an important element necessary for humans and other living organisms, which included numerous enzymes and proteins. Copper is used in the manufacturing of electrical wiring, fittings, valves, pipes, cooking utensils, coins, and building materials. On other hand copper compounds are utilized in insecticides, algaecides, fungicides, wood preservatives, azo dye synthesis, electroplating industry, engraving, lithography, pyrotechnics, and petroleum refining plants. However, copper is recognized as one of heavy metals that generates serious environmental hazards, wastewaters from metal finishing, weaving, and electronics industries may contain copper with concentration up to 500 mg/l. Based on the worldwide environmental regulations, this level of concentration is higher than the permitted level and treatment of these wastewaters must be achieved before being discharged into the environment [1]. The allowable limit of copper in sources of drinking water like rivers is in the range of 1.5 to 2 mg/L based on the European Union restrictions, so it is preferred to discharge the effluents within this limit [2]. Removal of copper from waste streams has been achieved by several methods comprising adsorption, chemical precipitation, ion-exchange, biosorption, electrodialysis, reverse osmosis, membrane separation based on ion-exchange, and electrochemical deposition [3]. Some of these methods are verified to be efficient in copper removal, however they have no ability to recover the valuable heavy metals that can the again. For example, a large amount of precipitated sludge is generated in the chemical precipitation technique which needs further treatment. Ion exchange and reverse osmosis techniques have limited applications because of the requirements of high material and operational costs. Electrochemical approach as a dramatic alternative to the well-known techniques offers electrochemical reactors that used electrochemical reduction reactions as a principal approach for removal of heavy metals ions from wastewater, where these metals are electrodeposited at the electrode surface as solid metallic deposits when the

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effluents flow through the electrochemical reactor hence the possibility of reusing them from the main process. Herein, adding chemicals is not required leading to facilitate of water reuse [4-6]. The electrochemical method is considered as a clean, environmentally engaging technology since the major reaction reagent is the electron. Economically, electrochemical method is valuable due to the lower energy consumption in comparing with the well-known techniques [5]. Besides, applying the automatization in controlling current during the electrodeposition process results in lowering the workload requirements [7]. Removal of copper from solutions have been attempted by several researchers using various cell designs including parallel plate electrochemical reactors [8-10], and packed bed electrochemical reactors [11-17] where some degrees of achievements and improvements have been achieved. In these works, two-dimensional electrode cells were found to be suffered extreme performance constraint, which was observed clearly as concentration limits of the effluents be stiffer. Porous, packed bed electrodes offer higher specific surface area which permits achieving higher removal rates of metal ion even at more dilute effluents [18]. To ensure efficient applying of the electrochemical method for diluted effluent treatment, the electrochemical reactors should have as possible as the higher value of the product of mass-transfer coefficient and specific surface area of the cathode which in turn improve the space time yield of the reactor. This aim can be accomplished by using packed bed rotating cylinder electrodes which have been recognized as an efficient type of electrochemical reactor that used for heavy metals removal [19]. This kind of electrochemical reactor has features not engaged by other reactors, for instance, the possibility of operating at continuous mode and a simple operable compact design [20, 19]. It can be run at concentration limit from 1 ppm to 1000 ppm, where the higher value resembles the concentration of most heavy metals in different industrial effluents while lower value matches the legislation constraints [21]. Heavy metals removal was carried out formerly by using rotating cylinder electrodes with a packed bed in the form of woven wire meshes and reticulated vitreous carbon (RVC) [22, 13, 23]. Previous studies showed that the packed bed rotating cylinder electrode of woven wire meshes when compared with the other types of packed bed rotating cylinder electrodes, has a value of mass transport coefficient greater approximately by three times than those observed in smooth rotating electrode as a result of higher turbulence-promoting action of the meshes [24], however, the electrode thickness should be kept small for assuring the whole bed to be work under limiting current conditions [25]. Tubular packed bed of woven screens cylinder electrode is one of packed bed rotating cylinder electrodes that not be used before as a packed bed rotating cylinder electrode for heavy metals removal [26]. In this configuration, the cathode was constructed from a number of coaxial closely packed layers of vertical screen cylinders. This type of rotating cylinder electrode has high turbulence-promoting action due to its high surface area per unit volume. Abdel-Aziz [26] studied the mass transfer in this type and found that this type has a value of volumetric mass transfer coefficient greater than that obtained at smooth rotating cylinder. Therefore, the major purpose of the present research is to examine the performance of a modified design of this type of rotating cylinder electrode for copper removal. The modified tubular packed bed rotating electrode is composed of a stainless steel perforated hollow cylinder which used as a current feeder where continuous layers of stainless steel screens are winded around it and bounded by two sleeves. This new configuration help in using high rotation speed hence higher turbulence action can be achieved. Besides, this configuration could be easily scaled–up to the industrial scale. The stated novelty of the present work is based on the using of tubular packed bed woven screen rotating cylinder electrode as a packed bed rotating electrode for copper removal.

In previous studies, the removal of heavy metals by electrochemical deposition method was studied utilizing a well-known one-factor-at-a-time method (OFAT). This method changes only one variable at a time whereas keeping others fixed. However, the interactions of the variables couldn't be specified from OFAT runs. The designed experiment method is a more effective method than OFAT method for evaluating the effect of two or more variables on the response of the process under study as well as their interactions. Lower resources (experiments, time, and materials) are needed by adopting the designed experiment technique to get the desired information. Besides, the evaluation of the effects of each variable is more accurate by adopting the designed experiment technique [27]. Response surface methodology (RSM) is a vital subject in the statistical design of experiments. It was used efficiently in different processes for wastewater treatment such as adsorption [28], disinfection of chlorine [29], electrocoagulation [30], Fenton-related process [31], electrochemical oxidation [32], and heavy metals removal [22]. Response surface methodology used a group of statistical and mathematical techniques for modeling and analyzing many problems in which various variables affected the response of the process. The object of RSM is to assessment the relative effect of various affecting variables and finally obtaining the optimum conditions by upgrading this response [33]. Hence, the second aim of this research is to optimize the variables of copper removal process like initial metal concentration, current, and rotation speed for improving copper removal efficiency from simulated wastewater using a tubular packed bed woven screens electrode. As a method of optimization, Box-Behnken design (BBD) of the response surface methodology was applied in this study. We believe that this is the first work that uses an optimization approach by BBD for electrochemical removal of copper utilizing a tubular packed bed woven screens electrode where no previous works have been reported in this field.

2. Experimental work

2.1. Materials and system

The electrolysis runs were performed in a 0. 5 L Perspex electrolytic cell. The cathode (working electrode) was a rotating tubular packed bed electrode composed of 316 stainless steel woven screens wrapped around stainless hollow cylinder acting as a current feeder. The hollow cylinder current feeder was opened at the bottom and closed at the upper. It is perforated with a total of (15) holes with a diameter (6mm) distributed uniformly on the lateral surface of the cylinder. The cathode feeder has an outer diameter (35 mm), inner diameter (28mm) with total length (60 mm). The lower part of this feeder is jointed with a Teflon sleeve has diameter (50mm) and height (12 mm), while the upper part is jointed with a Teflon sleeve has diameter (50mm) and height (17 mm) in order to fix the wrapped woven screens sheets on the current feeder. The cathode current feeder was attached to the shaft of variable speed motor via a stainless steel rod (7 mm diameter and 100 mm length) fixed on the cathode feeder. The cathode has an apparent surface area of (117.81 cm^2) (50 mm diameter and 60 mm long). Outer graphite cylinder having dimensions (90 mm inside diameter, 5 mm thickness, and 90 mm long) and central graphite rod having dimensions (60 mm length and 20 mm diameter) were used as anode (counter electrode). For ensuring a uniform primary current distribution, the three electrodes (cathode, outer anode, and inside anode) were concentric in the cell body. Figure 1 displays the schematic diagram of the experimental setup.

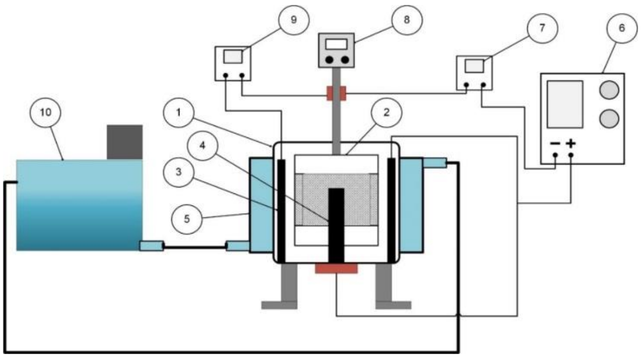


Figure 1: Schematic diagram of the experimental setup:1) cell body, 2) cathode, 3) outside anode, 4) inside anode, 5) jacket, 6) power supply, 7) Ammeter, 8) electrical motor,9) voltmeter,10) water bath circulator

Before starting any run, the cathode was washed with (1M) nitric acid solution in an ultrasound cleaner for removing copper deposits of the previous run then rinsed again thoroughly by double-distilled water. The galvanostatic copper deposition was conducted by using power Supply-model TP-1305EC, 30V / 5A. Stainless steel screens having mesh numbers 30 and 60 were used. The properties of these screens are presented in Table 1. Screen porosity (ε) was evaluated by determining the screen weight /area density and applying Eq. 1, then screen specific surface area(s) was computed based on Eq.2 [34]:

$$\varepsilon = 1 - \frac{m_s}{\rho_s l a_s} \quad (1)$$

$$s = (1-\varepsilon) r \quad (2)$$

where (r) is the surface to volume ratio of the screen wire equal to (4/d), (m_s/a_s) is the weight /area density, (ρ_s) is the density of stainless steel 316-AISI equal to 8.027gm/cm^3 [35], (*l*) is the screen thickness equal to 2d.

The woven type of the screen was identified by using Olympus BX51M with DP70 digital camera system whereas a digital caliper was used to measure wire diameter (d).

Table 1. Screen properties

Mesh number (wire/inch)	30	60
Type of woven	Plain square	Full twill
d, cm	0.030	0.020
(m_s/a_s), g/cm^2	0.1237	0.1291
ε	0.7146	0.6345
s, cm^{-1}	38.06	73.1

Copper sulfate ($CuSO_4$) is used as a source of copper ion while sodium sulfate (Na_2SO_4) was used as a supporting electrolyte. All chemicals were of reagent grade. Doubly distilled water was used for preparing electrolytic solutions containing copper ions dissolved in 0.5M Na_2SO_4 at concentrations (100, 200, 300, 400, and 500 ppm). The final pH of electrolytic solutions was 2 adjusted by using (1M) H_2SO_4 or (1M) NaOH. All runs proceeded at a fixed temperature of 30±1°C.

The removal efficiency (RE, %) was computed according to the following equation [36] :

$$RE = \frac{C_i - C_f}{C_i} \times 100 \quad (3)$$

where C_i is the initial copper concentration, C_f is the final copper concentration after an interval of time (Δt).

Current efficiency (CE, %) is the ratio of the actual mass of copper ion electrodeposited on the cathode surface to the theoretical mass that could be electrodeposited according to Faraday's law, it can be determined according to the following equation [36] :

$$CE = \frac{100 z_i \cdot F \Delta m}{M_i \, I \, \Delta t} \quad (4)$$

where *F is* the Faraday constant (96487A s mol^{-1}); Δm is the mass of copper electrodeposited at period of time Δt (g); M_i is the molar mass of copper (63.546 g/mol), z_i is the number of electrons, I is the applied current (A) and Δt is the electrolysis time (s).

The specific energy required for operating the electrochemical reactor is the major item in evaluating the cost of any electrochemical process. It is defined as the energy required for producing or treating a certain amount of the substance on a molar, mass, or volume basis. Specific energy consumption (EC, kWh kg^{-1}) can be evaluated according to the following equation [36]:

$$EC = \frac{2.788 \times 10^{-4} E I \Delta t}{\Delta m} \quad (5)$$

where *E* is the voltage of cell (Volt).

2.2. Design of experiments

The relationship between a process response and its variables can be determined by applying a collection of mathematical and statistical techniques adopted by RSM [37]. In this study, the 3-level 3-factor Box–Behnken experimental design is implemented to verification and check the variables that influenced the removal of copper from simulated wastewater. Current (X1), rotation speed (X2), and initial copper ion concentration (X3) were taken as process variables, while the efficiency of copper removal was taken as a response. The scales of process variables were coded as -1 (low level), 0 (middle or central point) and 1 (high level) [38]. Table 2 illustrates the process variables with their chosen levels. Box–Behnken improves designs to get the suitable quadratic model with the required statistical properties by using only a part of the runs needed for a 3-level factorial. The number of runs (N) needed for performing of Box–Behnken design can be determined by the following equation [39]:

$$N = 2k(k-1) + cp \quad (6)$$

where k is the number of process variables and cp is the reiterated number of the central point.

Table 2, Process variables with their level for copper removal

Process variables	Levels in Box–Behnken design		
Coded levels	Low(-1)	Middle(0)	High (+1)
X1-AppliedCurrent(A)	0.5	1.5	2.5
X2-Rotation speed(rpm)	150	450	750
X3-Cu(II) concentration(ppm)	100	300	500

In this research, fifteen runs were conducted for evaluating the effects of the process variables on the copper removal efficiency. Table 3 illustrates the BBD proposed for the present research.

Table 3, Box- Behnken experimental design

Run	Blk	Coded value			Real value		
		X1	X2	X3	Current (A)	Rotation Speed (rpm)	Concentration (ppm)
1	1	-1	0	1	2.5	450	100
2	1	-1	-1	0	1.5	150	100
3	1	-1	0	-1	0.5	450	100
4	1	0	0	0	1.5	450	300
5	1	0	-1	1	2.5	150	300
6	1	1	-1	0	1.5	150	500
7	1	1	0	-1	0.5	450	500
8	1	0	0	0	1.5	450	300
9	1	0	0	0	1.5	450	300

A second order polynomial model can be adopted based on BBD were fitting the interaction terms with the experimental data can be described by the following equation [40]:

$$Y = a_0 + \sum a_i x_i + \sum a_{ii} x_i^2 + \sum a_{ij} x_i x_j \tag{7}$$

where Y represents the dependent variable (RE), i and j are the index numbers for patterns, a_0 is the intercept term, $x_1, x_2 \ldots x_k$ are the process variables (independent variables) in coded form. a_i is the first-order(linear) main effect, a_{ii} second-order main effect and a_{ij} is the interaction effect. Analysis of variance was performed then the regression coefficient (R^2) was estimated to confirm the goodness of model fit.

3. Results and discussion

3.1. Statistical analysis

The optimization of process variables and identification of the interaction among them were performed by conducting fifteen runs at different combinations of the process variables. Table 4 shows the values of the removal efficiency for each run. Current efficiency and specific energy consumption are also inserted in this Table. It is interesting to observe that copper removal efficiency was changed from 92.27 to 99.87%, current efficiency altered from 2.6912 to 62.625%, while the energy consumption was in the range of 3.115-137.349 Kwh/kg when adopting the experimental design.

Table 4, Experimental results of Box–Behnken design for copper removal

Run	Blocks	Real Value			RE%		E Volt	CE %	EC kWh/kg
		Conc. (ppm)	Rotation (rpm)	Current (A)	Actual	Predict			
1	1	100	450	2.5	99.70	99.88	4.37	2.69	137.35
2	1	100	150	1.5	96.80	96.89	3.59	4.36	69.77
3	1	100	450	0.5	96.10	96.23	2.70	12.97	17.60
4	1	300	450	1.5	97.10	97.13	3.53	13.11	22.81
5	1	300	150	2.5	97.60	97.33	4.09	7.90	43.82
6	1	500	150	1.5	92.27	92.66	3.25	20.76	13.26
7	1	500	450	0.5	92.80	92.62	2.31	62.63	3.12
8	1	300	450	1.5	97.10	97.13	3.53	13.11	22.81
9	1	300	450	1.5	97.10	97.13	3.53	13.11	22.81
10	1	300	750	2.5	99.87	100.1	4.74	8.09	49.55
11	1	100	750	1.5	99.25	98.86	3.65	4.47	69.08
12	1	500	750	1.5	97.70	97.61	3.46	21.98	13.33
13	1	300	150	0.5	92.32	92.11	2.60	37.38	5.89
14	1	300	750	0.5	96.00	96.27	2.35	38.87	5.11
15	1	500	450	2.5	98.14	98.01	3.64	13.25	23.24

Table 5, Analysis of variance for copper removal

Source	DF	Seq SS	Cr. (%)	Adj SS	Adj MS	F-Value	P-Value
Model	9	84.2003	98.94	84.200	9.3556	51.75	0.0
Linear	3	79.7751	93.74	79.775	26.5917	147.10	0.0
X1-Current(A)	1	40.9060	48.07	40.906	40.9060	226.29	0.0
X2-Rotation speed (rpm)	1	23.9086	28.09	23.908	23.9086	132.26	0.0
X3-Concentration (ppm)	1	14.9604	17.58	14.960	14.9604	82.76	0.000
Square	3	0.9512	1.12	0.9512	0.3171	1.75	0.272
X1*X1	1	0.2362	0.28	0.2362	0.2362	1.31	0.305
X2*X2	1	0.6350	0.75	0.6920	0.6920	3.83	0.108
X3*X3	1	0.0800	0.09	0.1410	0.1410	0.78	0.418
2-Way Interaction	3	3.4740	4.08	3.4740	1.1580	6.41	0.036
X2*X3	1	2.2201	2.61	2.2201	2.2201	12.28	0.017
X1*X3	1	0.7569	0.89	0.7569	0.7569	4.19	0.096
X1*X2	1	0.4970	0.58	0.4970	0.4970	2.75	0.158
Error	5	0.9038	1.06	0.9038	0.1808		
Lack-of-fit	3	0.6572	0.77	0.6572	0.2191	1.78	0.380
Pure error	2	0.2467	0.29	0.2467	0.1233		
Total	14	85.1041	100				
Model Summary	S	R-sq	R-sq(adj.)	PRESS	R-sq(pred.)		
	0.425	98.94%	97.03%	11.069	86.99%		

Minitab-17 software was used to analyze results of copper removal efficiency where an experimental relationship between copper removal efficiency and process variables was obtained and formulated by the following quadratic model of copper removal efficiency (RE) in term of coded units of process variables:

$$RE\% = 93.08 + 2.896\,X1 + 0.00813\,X2 - 0.01276\,X3 - 0.000005\,X3^2 - 0.000005\,X2^2 - 0.253\,X1^2 + 0.000012\,X3{*}X2 + 0.00217\,X3{*}X1 - 0.001175\,X2{*}X1 \quad (8)$$

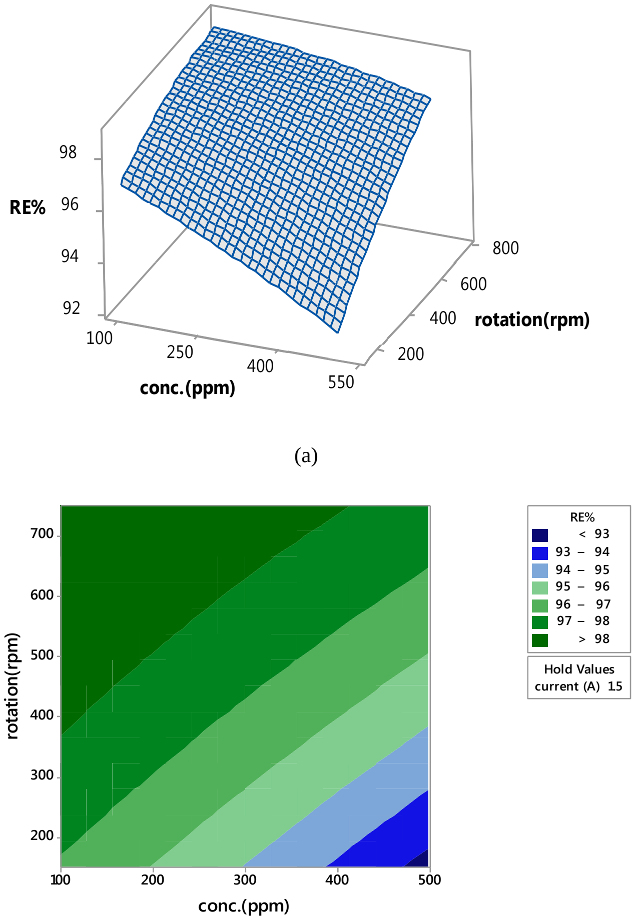
Eq.(8) shows how the removal efficiency is affected by the individual variables (linear and quadratic) or double interactions. The values of positive coefficients revealed that the removal efficiency increased with the increasing of the related factors of these coefficients within the tested range while values of negative coefficients revealed the opposite effect. As can be seen, concentration has a negative effect on the removal efficiency, while current and rotation speed were found to have a positive effect. The results showed that effects of interactions are not significant. The predicted values of the removal efficiency estimated from Eq.8 are also inserted in Table 4.

The Box-Behnken design adequacy was identified by using an analysis of variance (ANOVA). To test hypotheses on the parameters of the model, ANOVA divides the total variation in a set of data into individual parts supplemented with specific sources of variation [41]. The adequacy of the model in ANOVA analysis is recognized based on Fisher F-test and P-test. Most of the variation in the response can be illustrated by the regression equation if the value of Fisher becomes higher. P-value is used for evaluating whether F is large enough to signalize statistical significance. 95% of the variability of the model could be clarified when a P-value lower than 0.05 [42]. Table 5 illustrates ANOVA for the response surface model. In this table, degree of freedom (DF), the sum of the square (SeqSS), percentage contribution (Cr. %) for each parameter , adjusted sum of the square (Adj SS), adjusted mean of the square (Adj MS), F-value, and P-value were evaluated. F-value of 51.57 and P-value of 0.0001were obtained which elucidating high significance for the regression model. The multiple correlation coefficient of the model was 98.94% conforming to the regression is statistically significant and only 1.06 % of the total variations are not confirmed by the model. The adjusted multiple correlation coefficient (adj. R^2 = 97.03%) and the predicted multiple correlation coefficient (pred. R^2 = 86.99%) were compatible with this model.

Results of ANOVA showed that percent of the contribution of the current is 48.07% which means that the current has the main effect on copper removal efficiency. Rotation speed and initial copper concentration have miner effects. The linear term has the main percent of contribution in the model with 93.74% followed by the interaction between the input variables with a contribution of 4.08%while the square has a small contribution (1.12%) which could be ignored. The results assure that current is the most significant factor.

3.2. Effect of process variables on the copper removal efficiency

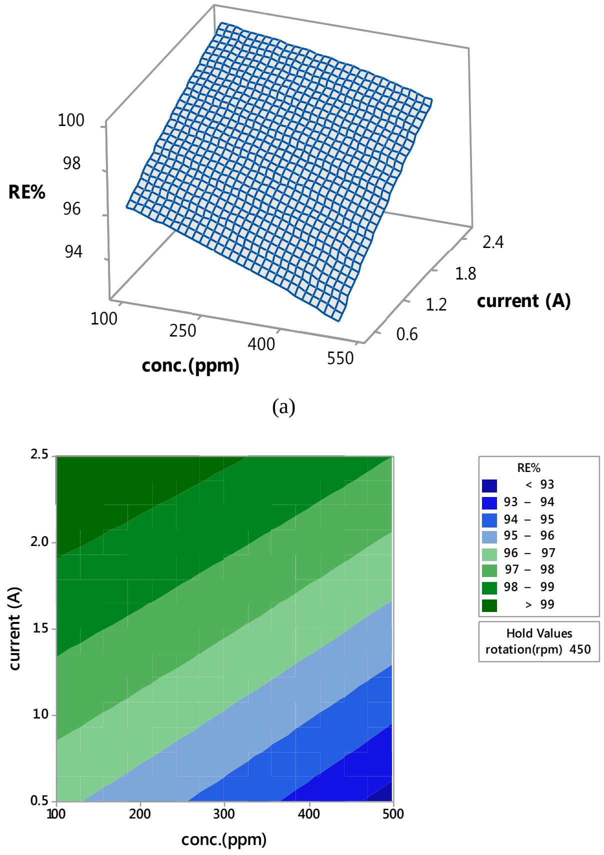
Figures (2-a, 2-b) show the effect of the initial copper concentration on copper removal efficiency for various values of rotation speeds (150, 300, 450, 600, and 750 rpm) at constant current (1.5 A) with mesh no. of 30. Figure 2-a represents the response surface plot while figure2-b shows the corresponding contour plot. From the surface plot, it was observed that, at a rotation speed of 150 rpm, a decrease in removal efficiency occurs as the initial copper concentration increased. However, a slight change in the removal efficiency happened as the rotation speed approach to 750 rpm. At the concentration of 500ppm, the results show an increase in copper removal efficiency with increasing rotation speed. However, at concentration of 100ppm, a slightly change in the removal efficiency was occurred with increasing rotation speed. The corresponding contour plot confirms that a maximum value of copper removal efficiency lies in a small area in which the rotation speed ranged between 500-750rpm and copper ion concentration between 100-200ppm. The effect of the current on copper removal efficiency for different initial copper concentrations (100, 200, 300, 400, and 500 ppm) at constant rotation speed of 450 rpm with mesh no. of 30 is shown in Figures (3-a, 3-b). The response surface plot (3-a) shows that currently has an important effect on copper removal efficiency where it increases quickly as the current raised up to 2.5 A. While removal efficiency slightly decreased with increasing concentration. The corresponding contour plot(3-b) confirms that a maximum value of copper removal efficiency lies in a small area in which the current ranged between 2-2.5 A and copper ion concentration between 100-200ppm.



(a)

(b)

Figure 2: Response surface plot (a) and contour plot (b) showing the effect of rotation and initial concentration of copper on the copper removal efficiency



(a)

(b)

Figure 3: Response surface plot (a) and contour plot (b) showing the effect of the current and initial concentration of copper on the copper removal efficiency

3.3. The optimization and confirmation test

Numerical optimization of the software is applied to get the precise point that maximized the desirability function (DF). The desired goal was chosen by adjusting the weight or importance that could change the characteristics of the aim. Five options for the aim fields for response were selected: maximum, minimum, target, within range, and none. In the present work, the aim is to get higher removal efficiency of copper so the 'maximum' field with corresponding 'weight'1.0 was chosen. 92.27% was taken as the lowest limit for the removal efficiency while 99.87% was taken as the upper limit. Under these settings and boundaries, the optimization procedure was conducted and the results are displayed in Table 6 with the desirability function of (1). Results of optimization recommended using the current of 2.5A, a rotation speed of 750 rpm, and an initial copper concentration of 205.05 to get higher removal efficiency of 100.3%.

Two experiments at the optimum values of the process parameters were performed to confirm the results of optimization. 205 ppm was taken as nearly the value of the initial copper concentration resulted from optimization. The results are displayed in Table 7. After 30 min of the electrolysis, the removal efficiency of 99.127% was achieved which is incompatible with the range of the optimum value getting from optimization analysis with desirability function of (1) (Table 6). Therefore adopting Box–Behnken design combined with desirability function is successful and efficient in optimizing copper removal using a tubular packed bed of woven screens rotating electrode.Reade et al. [23] investigated the potentiostatic removal of copper from acid sulfate solutions using reticulated vitreous carbon (RVC) rotating cylinder electrode. They found that an initial copper concentration of 63.5ppm could be reduced to <0.1ppm in approximately 60 min using a 100 ppi RVC at electrode potential of −500mV vs SCE. The present work gives the same removal efficiency starting from an initial copper concentration of 205 ppm at half interval time under galvanostatic operation mode (constant current) which is an indication of the good performance of the present modified rotating cylinder electrode, moreover, the galvanostatic operation mode is the preferred mode at the industrial scale. Other previous works that used rotating packed bed cylinder electrode were operated at single-pass flow mode of operation not batch mode [13].

Table 7 shows that current efficiency was 5.2% which means that most of the current is consumed for hydrogen evolution as a side reaction. This lower current efficiency is expected since the concentration of copper very low (205 ppm) and pH of the solution is 2. Previous works stated that hydrogen ions discharge as a side reaction is strongly competitive to the electrodeposition of copper ions on the surface of cathode as the acidity of the solution is increased [43]. Of course operating at pH higher than 2 will offer superior removal of copper by electrodeposition at higher current efficiency. This can be achieved with generous caution since copper could be precipitated as hydroxide if the solution pH is greater than the value of pH for precipitation as approved by theoretical solubility of copper hydroxide diagram[44].Therefore most of previous works operated at pH=2 [23, 13]. Although the literature reports some values of current efficiency higher than we found at galvanostatic mode of operation, the present rotating cylinder electrode used in this study has shown very satisfactory performance in removal of copper. In addition the hydrogen evolution can be utilized as a chemical source for other industrial applications when a divided cell configuration is adopted at the industrial scale, hence another benefit from the present research can be obtained.

3.4. Effect of mesh number

To investigate the effect of mesh no. on the removal efficiency, two runs were performed at the optimum conditions using two mesh no. 30 and 60. The concentration profile with time for different mesh no. is shown in Fig 4. It is clear there is an insignificant effect of two mesh numbers on the removal efficiency where the same concentration profiles were observed. This behavior is in good agreement with our previous research [22] in which cadmium removal by using a spiral-wound woven wire mesh packed bed rotating cylinder electrode was studied where cadmium removal efficiency was found to be not significantly changed with increasing of mesh number.

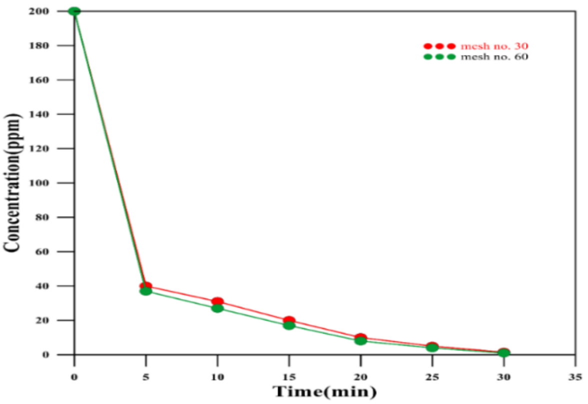


Figure 4, Concentration versus time for two sizes of mesh no

Table 6. The optimum values of process parameters for maximizing copper removal efficiency

Response	**Goal**	**Lower**	**Target**	**Upper**		**Weight**	**Importance**
RE (%)	**Maximum**	92.27	99.87	100		1	1
Solution:Parameters			**Results**				
X1 (A)	**X2 (rpm)**	**X3 (ppm)**	**RE (%) Fit**	**D_F**	**SE Fit**	**95% CI**	**95% PI**
2.5	750	205.05	100.13	1.0	0.39	(99.123;101.137)	(98.644;101.62)

Table 7. Confirmation of the optimum conditions for copper removal efficiency

Run	**Run**	**X1 (A)**	**X2 (rpm)**	**X3 (ppm)**	**E (Volt)**	**R E(%) at 30 min**		**CE (%)**	**EC (Kwhkg-1)**	**RE(%) at 40 min**
						actual	**average**			
1	1	2.5	750	205	2.7	99.25	4. 99.12	5.2	77	5. 100
2	2	2.5	750	205	2.8	99				

4. Conclusions

It was established that copper removal from a simulated wastewater solution could be performed successfully in a rotating tubular packed bed of woven screens electrode as a cathode in a batch electrochemical reactor. RMS methodology is applied effectively for optimizing the process parameters and finding out the optimum levels of these parameters for copper removal which maximized the removal efficiency. Based on RSM analysis, it can be concluded that currently has the largest effect on the efficiency of electrochemical copper removal in comparison with the other parameters. The optimal values obtained from the optimization were Cu (II) initial concentration of 205 ppm, current of 2.5A, and rotation speed of 750 rpm. Under these conditions, it could be possible to reduce Cu (II) concentration from 205 ppm to less than 2 ppm (RE=99.12%) at electrolysis time of 30 min and a complete removal was obtained at 40 min. Therefore, an additional benefit of the present system was gained represented by achieving complete removal and recovery of copper.

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REFERENCES

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[1] K. Dermentzis, A. Davidis, D. Papadopoulou, A. Christoforidis, K. Ouzounis .Journal of Engineering Science and Technology Review, 2 :131,2009.

[2] F. Imre-Lucaci, PhD thesis, “Babe¸s-Bolyai,” University Cluj–Napoca, Faculty of Chemistry and Chemical Engineering, 2011.

[3] C. Ahmed Basha, N.S. Bhadrinarayana, N. Anantharaman, Meera Sheriffa Begum, K.M. J. Hazard. Mater. 152 ,71–78, 2008.

[4] F. Fu, Q. Wang, “Removal of metal ions from wastewaters,”: A review. J. Environ. Manage. 92, 407-418, 2011.

[5] K. Juttner, U. Galla, H. Schmieeder, “Electrochemical approaches to environmental problems in the process industry,” Electrochim Acta. 45, 2575–2594, 2000 .

[6] D. Pletcher, F. C. Walsh, “Industrial Electrochemistry,” Chapman and Hall, New York, 1990.

[7] G. A. Tonini, F. R. Martins, P. F. De Almeida Prado, L. A. Martins Ruotolo, “Box–Behnken factorial design study of the variables affecting metal electrodeposition in membrane less fluidized bed electrodes,” J. Chem. Technol. Biotechnol. 88, 800–807, 2013.

[8] C. Solisio, M. Panizza, P. Paganelli, G. Cerisola, “Electrochemical remediation of copper (II) from an industrial effluent Part I: monopolar plate electrodes,” Resour. Conserv. Recycl. 26, 115–124, 1999.

[9] S. Chellammal, S. Raghu, P. Kalaiselvi, G. Subramanian, “Electrolytic recovery of dilute copper from a mixed industrial effluent of high strength,” CODJ Hazard Mater. 180 (1–3) , 2010.

[10] C. Ahmed Basha, R. Saravanathamizhanb, V. Nandakumarc, , K. Chitrad, Chang Woo Lee, “Copper recovery and simultaneous COD removal from copper phthalocyanine dye effluent using bipolar disc reactor,” Chem. Eng. Res. Design (91) 552–559, 2013.

[11] A.K.P. Chu, M. Fleischmann, G.J. Hills, “Packed bed electrodes. I. The electrochemical extraction of copper ions from dilute aqueous solutions J. App,” Electrochem. 4, 323–330, 1974

[12] G. Issabayeva, M.K. Aroua, N. Sulaiman, “Electrodeposition of copper and lead on palm shell activated carbon in a flow-through electrolytic cell,” Desalination 194, 1–3, 2006.

[13] J. M. Grau, J. M. Bisang, “Effluent treatment using a bipolar electrochemical reactor with rotating cylinder electrodes of woven wire meshes,” J. Chem. Technol. Biotechnol. 84, 1084–1089, 2009.

[14] S.H. Chang, K.S. Wang, P.I. Hu, I.C. Lui, “Rapid recovery of dilute copper from a simulated Cu–SDS solution with low-cost steel wool cathode reactor,” J. Hazard. Mater. 163, 544–549, 2009.

[15] C. Ahmed Basha, T. Somasundaramb, T. Kannadasanb, Chang Woo Lee, “Heavy metals removal from copper smelting effluent using electrochemical filter press cells,” Chem. Eng. J. 171 , 563–571, 2011.

[16] I.A. Khattab, M.F. Shaffei, N.A. Shaaban, H.S. Hussein, S.S. Abd El-Rehim, “Electrochemical removal of copper ions from dilute solutions using packed bed electrode,” Part I. Egyptian Journal of Petroleum , 22, 199–203, 2013.

[17] I.A. Khattab, M.F. Shaffei, N.A. Shaaban, H.S. Hussein, S.S. Abd El-Rehim, “Electrochemical removal of copper ions from dilute solutions using packed bed electrode,” Part II. Egyptian Journal of Petroleum, 22, 205-210, 2013.

[18] C.A.C. Sequeira, “Environmentally Oriented Electrochemistry,” Elsevier Science, Amsterdam, 1992.

[19] F.C. Walsh, “The role of the rotating cylinder electrode reactor in metal ion removal,” in: Electrochemistry for a Cleaner Environment, ed by Genders D and Weinberg N. The Electrosynthesis Company Inc, New York. 101-159, 1992.

[20] D. R. Gabe, “The rotating cylinder electrode,” J. Appl. Electrochem. 4, 91-108, 1974.

[21] J. St-Pierre, N. Masse, E. Friechette, M. Bergeron, “Zinc removal from dilute solutions using a rotating cylinder electrode reactor,” J. Appl. Electrochem. 26,369- 377, 1996.

[22] A. H. Abbar, R. H. Salman, A. S. Abbas, “Cadmium removal using a spiral-wound woven wire meshes packed bed rotating cylinder electrode,” Environmental Technology & Innovation 13, 233–243, 2019.

[23] G. W. Reade, A. H. Nahle, P. Bond, J. M. Friedrich, F. C. Walsh, “Removal of cupric ions from acidic sulfate solution using reticulated vitreous carbon rotating cylinder electrodes,” J. Chem. Technol. Biotechnol. 79, 935–945, 2004.

[24] A. H. Abbar, R. H. Salman, A. S. Abbas, “Studies of mass transfer at a spiral-wound woven wire mesh rotating cylinder electrode,” Chem. Eng. Process. - Process Intensif. 127, 10–16, 2018.

[25] G. Kreysa, R. Brandner, In: Modern Concepts in Electrochemical Reactor Design, Extended Abstracts of the 31st ISE Meeting. Venice, Italy, 2: H8, 1980.

[26] M.H. Abdel-Aziz, I. Nirdosh, G.H. Sedahmed, “Mass transfer at a rotating tubular packed bed of woven screens in relation to electrochemical and catalytic reactor design,” International Journal of Heat and Mass Transfer 90, 427–438, 2015.

[27] L.-C. Cheng, W.-L. Chou, C.-P. Chang, Y.-M. Kuo, C.-T. Wang, “Application of response surface methodology for electrochemical destruction of cyanide,” Int. J. Phys. Sci. 7(44), 5870-5877, 2012.

[28] H. Sabah, T. Thouraya, H. Melek, M. Nadia, “Application of Response Surface Methodology for Optimization of Cadmium Ion Removal from an Aqueous Solution by Eggshell Powder,” Chem. Res. Chin. Univ.34, 302-310, 2018.

[29] M. Umar, H. A. Aziz, M.S. Yusoff, “Assessing the chlorine disinfection of landfill leachate and optimization by response surface methodology (RSM),” Desalination 274,278-283, 2011.

[30] D. Prabhakaran, C. A. Basha, T. Kannadasan, P. Aravinthan, “Removal of hydroquinone from water by electrocoagulation using flow cell and optimization by response surface methodology,” J Environ. Sci. Health. Part. A. 5,400-412, 2010.

[31] A. Khataee, M. Zarei, L. Moradkhannejhad, “Application of response surface methodology for optimization of azo dye removal by oxalate catalyzed photoelectro-Fenton process using carbon nanotube-PTFE cathode,” Desalination 258,112-119, 2010.

[32] B.K. Körbahti, “Response surface optimization of electrochemical treatment of textile dye wastewater,” J. Hazard. Mater. 145,277-286, 2007.

[33] K. Thirugnanasambandham, V. Sivakumar, M. J. Prakash, “Treatment of egg processing industry effluent using chitosan as an adsorbent,” J. Serb. Chem. Soc. 79(6), 743-757, 2014.

[34] R. E. Sioda, “Mass transfer problems in electrolysis with flowing solution on single and stacked screens,” J. Electroanal. Chem.70 (1),49-54, 1976.

[35] Green, D., Perry, R., 2008. Perry’s Chemical Engineers’ Handbook, 8thed. McGraw-Hill, New York.

[36] A. H. Sulaymon, , Mohammed, A. H. Abbar, “Cadmium removal from simulated chloride wastewater using a novel flow-by fixed bed electrochemical reactor: Taguchi approach,” Desalination Water Treat. 74, 197–206, 2017.

[37] M.A. Bezerra, R.E. Santelli, E.P. Oliveira, L.S. Villar, L.A. Escaleira, "Response surface methodology (RSM) as a tool for optimization in analytical chemistry," Talanta 76(5), 965-77, 2008.

[38] M. Evans, "Optimization of Manufacturing Processes: A Response Surface Approach," Carlton House Terrace, London, 2003.

[39] Y.,-D. Chen, W.-Q. Chen, B. Huang, M.-J. Huang, "Process optimization of K2C2O4-activated carbon from kenaf core using Box–Behnken design," Chem. Eng. Res. Des. 91(9), 1783-1789, 2013.

[40] K. Yetilmezsoy, S. Demirel, R.J. Vanderbei, "Response surface modeling of Pb(II) removal from aqueous solution by Pistacia vera L.: Box–Behnken experimental design," J. Hazard. Mater.171 (1–3), 551-562, 2009.

.

[41] L. Huiping, Z. Guoqun, N. Shanting, L. Yiguo, "Technologic parameter optimization of gas quenching process using response surface method," Comput. Mater. Sci. 38(3), 561–570, 2007.

[42] J. Seguroleat, N.S. Allen, M. Edge, A.M. Mahon, "Design of eutectic photoinitiator blends for UV/curable acrylated printing inks and coating," Prog. Org. Coat. 37, 23–37, 1999.

[43] M. F. Alebrahim, I.A. Khattab, A. O. Sharif, "Electrodeposition of copper from a copper sulfate solution using a packed-bed continuous-recirculation flow reactor at high applied electric current," Egyptian Journal of Petroleum 24, 325–331, 2015.

[44] D. M. Ayres, A. P. Davis, G. M. Paul, "Removing Heavy Metals from Wastewater," University of Maryland, Engineering Research Center Report, (p. 6), 1994