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Cadmium removal from simulated chloride wastewater using a novel flow-by fixed bed electrochemical reactor: Taguchi approach

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ABSTRACT

Taguchi parameter design approach was used for optimization of process parameters for cadmium removal from chloride simulated wastewater using a flow-by fixed bed electrochemical reactor composed of a vertical stack of stainless steel screens. The studied process parameters were metal ion concentration, current, flow rate, and mesh number of screen. Removal and current efficiencies combined with energy consumption were considered as responses for optimizing of metal removal. An orthogonal array L_9, the signal-to-noise ratio and the analysis of variance (ANOVA) were used to analyze the effect of the selected parameters and their levels. ANOVA results indicate that only the current has the major effect on the removal efficiency (RE) while concentration has the major effect on both current efficiency (CE) and energy consumption (EC) followed by current. Flow rate and screen mesh number have a lower contribution on the performance of cadmium removal. The optimum values of the control factors were: [Cd] = 200 ppm, 1.72 A, 5 L/min, and 30 in^{-1} in which a higher RE of 99.0% was obtained at CE of 28%, EC of 9.0 kWh/kg, and electrolysis time of 50 min. The new electrochemical reactor was found to be more efficient in the removal of cadmium when compared with the traditional fixed bed electrochemical reactors because of the high rate of mass transfer that was observed.

Keywords: Heavy metals; Electrochemical reactor; Cadmium; Flow-by electrode; Taguchi method

1. Introduction

Heavy metals released into the environment have been increasing continuously as a result of industrial activities such as electroplating, photographic development, printed circuit board production, and battery technology [1]. The release of large quantities of these metals into the natural environment such as irrigation of agricultural fields by using sewages has resulted in a number of environmental problems due to their non-biodegradability and persistence [2]. Therefore, they can accumulate in the environment elements such as in food chain, and thus may pose a significant danger to human health [3]. Cadmium is one of the most important toxic metals which should be removed before discharging to environment. Cadmium has been classified by U.S. Environmental Protection Agency as a probable human carcinogen. Chronic exposure of cadmium results in kidney dysfunction and high levels of exposure will result in death [4].

A number of techniques have been used to remove heavy metals from wastewater effluents; including chemical precipitation [5], electrodialysis [6], ion-exchange process [7,8], adsorption onto activated carbon [9,10], low cost adsorbents such as kaolin, bentonite, blast furnace slag, and fly ash [11],

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ion imprinted polymer [12,13], organic-based ligand precipitation [14], and membrane and reverse osmosis processes [15]. The industrial utilization of these methods has been found to be limited, because of the high capital and operating costs and/or the ineffectiveness in meeting stringent effluent standards [16].

Electrochemical cleaning technology by cathodic deposition offers an efficient means of controlling pollution through the removal of transition and heavy metals by redox reactions, without the disadvantages of conventional treatments. The inherent advantage of this technology is its environmental compatibility due to the fact that the main reagent, the electron, is a 'clean reagent' [17,18]. The increasing use of electrochemical technologies in the environmental treatment is due to the utilization of porous materials as a three-dimensional (3-D) electrode in the design of electrochemical reactors [19]. One of the main advantages of this kind of electrode derives from the fact that it can provide a high specific surface area as well as a high mass transfer rate. Two principal configurations for the 3-D electrodes have been developed: the flow-through configuration, where fluid flow and current are parallel; and the flow-by configuration, where the fluid flows perpendicularly to the current [20]. Unfortunately the flow-through porous electrode has met with a limited success on the commercial scale in view of the non-uniformity of current and potential distribution, poor selectivity, and low conversion per pass [21]. To avoid these shortcomings, attention has been directed to the flow-by electrode which has other advantages such as the possibility of using it in the form of a divided or undivided filter press type cell [22–24].

Several types of flow-by electrode have been proposed, for example, carbon or metal particles [25,26], metallic or metal plated foams and felts [27,28], and reticulated vitreous carbon (RVC) [29]. The main drawback of these electrodes, however, is the fact that continuous metal deposition leads to clogging of the pores by the deposited metal. In addition, they suffer from the high pressure drop. Besides, flow-by fixed bed electrodes made of small particles, metal felt and metal foam may entrap gas bubbles (H_2) which are likely to evolve simultaneously with the main reaction from dilute solutions with a consequent increase in cell resistance and electrical EC [30]. The use of screens and expanded metals in building 3-D electrodes offers many advantages, such as high specific area, high turbulence-promoting ability, high porosity and relatively low pressure drop, ease of coating with a catalyst, and ready availability at modest cost [31]. In addition, they present a rigid structure and are relatively easy to construct.

A variety of test works utilizing metallic nets, grids, or screens have been reported [32–39]. Most of these works concerned with the theoretical and experimental aspects of current and potential distributions in addition to the mass transfer correlations that describe the electrochemical systems, however, few works have been reported on studying the performance of these reactors in removal of heavy metal ions with economical and optimal approach [40–42]. On the other hand, for metals like cadmium, for example, the electrodeposition process must be optimized in order to make the electrochemical technology competitive with the precipitation process. In this sense, the electrochemical technology needs to be well understood under the kinetic and mass transfer aspects in order to ensure adequate reactor design and to maximize the reaction rate and CE with consequent minimization of EC [43].

To evaluate the performance of the removal process, Taguchi method was selected as a statistical technique once it allows the main effects to be estimated with a minimum number of experiments where a design plan based on the orthogonal arrays was introduced by Taguchi. Through the application of Taguchi's signal-to-noise (S/N) ratio, the optimal operational conditions from an experiment can be determined [44].

The main objective of this work is to optimize process parameters for cadmium removal from simulated chloride wastewater using a flow-by fixed bed electrochemical reactor composed of a vertical stack of stainless steel screens. The choice of stainless steel as a cathode material is based on the observation of previous works that stainless steel has been proved to be very effective as cathode for metal removal from wastewaters [45,46]. This material showed a good stability as cathode and could also be anodically polarized during its regeneration process without damage. Furthermore, stainless steel is an inexpensive material when compared with graphite felt or RVC which has been extensively used in wastewaters treatment. The choice of a vertical orientation of stack is based on the observations of Storck et al. [22] and Mobarak et al. [35] that this geometry is better for scale-up as the current and potential distribution are uniform. The effects of metal ion concentration, current (*I*), flow rate (*Q*), and mesh number of screen (*N*), on the removal and current efficiencies in addition to the EC were investigated.

2. Experimental work

2.1. Materials and system

The system for cadmium removal consists of an electrochemical cell, two 5-L capacity Perspex reservoirs for the catholyte and the anolyte, two magnetic recirculation pumps (EHEIM Kreiselpumpe, W. Germany), and two sets of flowmeters, with a flow range from 0 to 8 L/min, used for controlling the catholyte and analyte flow rates. A scheme of the system is shown in Fig. 1. This arrangement enabled the recirculation of anolyte and catholyte in two separated loop through the reactor in a batch recycle mode. The experiments were conducted at ambient temperature and under galvanostatic conditions. The current control was achieved by using a constant current source (Power Supply—model UNI-T: UTP3315TF-L). Cell current and potential were measured using multimeters type Pro'skit MT-1210 and MT-1280, respectively.

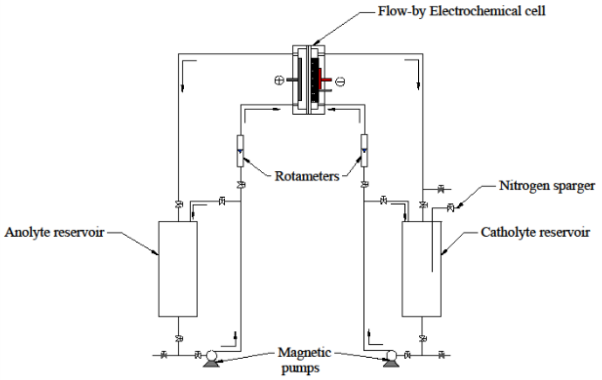

Fig. 1. Schematic view of the batch recirculation electrochemical system.

The flow-by reactor in this study is designed based on the criteria established by Risch and Newman [47]. According to their criteria, a flow-by reactor with an aspect ratio of the electrode length to thickness, $L/d > 5$, will produce a maximum processing rate than a flow-through configuration. The aspect ratio of the present cell is 33.33. The electrolysis cell, which is shown in Fig. 2, is basically a rectangular flow channel constructed from two machined blocks of polytetrafluoroethylene (PTFE). The first is the cathodic chamber having external dimensions (30 × 14 × 2.5 cm) while the second is the anodic chamber with dimensions (30 × 14 × 3.5 cm). The anodic chamber has two cavities; internal (10 × 10 × 2.2 cm) in which graphite block (10 × 10 × 2 cm) working as anode was fixed and external (24 × 10 × 0.5 cm) in which the anolyte is flowing over the anode upward. The graphite anode has carbon content (99%), ash content ≤ 0.5%, resistivity ≤ 10.0 μΩ, and bulk density of 1.72 g/cm^3 (Topfly Material Co. Ltd., China). It was grooved lengthwise to increase its surface area. The cathode chamber has also two cavities; internal (10 × 10 × 0.6 cm) in which copper plate (10 × 10 × 0.5 cm) working as current feeder was fixed, and external (24 × 10 × 0.3 cm) in which the catholyte passes through two stacks of screens: the first composed of seven polypropylene meshes with mesh number of 30 in^{-1} working as calming zone while the second consisted of five stainless steel screens (10 × 10 cm) working as a flow-by electrode. The current feed to the electrodes was provided by screw connectors through the walls of the cell. A saturated calomel reference electrode within a Teflon Luggin capillary passes the cathode chamber through a 3-mm hole drilled at the back near the copper plate. The anodic and cathodic chambers were separated from each other by a cationic membrane (IONIC-64LMR) which was supported on both sides with 2 mm thickness PTFE perforated plates. The cell was held vertically and the electrolytes were circulated upward.

Chemicals used were $CdCl_2.2H_2O$, NaCl, and boric acid. All were of reagent grade. The electrolytic solutions were freshly prepared with doubly distilled water. Solutions containing metallic ion were prepared in such a way that the metallic concentration was between 200 and 50 mg/L in supporting electrolyte consisting of 0.5 M NaCl and 0.1 M H_3BO_3 with final pH of 5 adjusted by using either 1 M NaOH or 1 M HCl. It is important to point out that such solutions were prepared trying to resemble the amount of metallic ions and pH of a typical composition contained in an effluent generated by a plating industry [48]. The catholyte reservoir was furnished by 4 L of electrolytic solution containing the cadmium ion while the anolyte reservoir was furnished with the supporting electrolyte only at the same volume. The catholyte was purged with high purity nitrogen (99.999%) for 30 min, while a nitrogen atmosphere was maintained over the catholyte during the experiment to prevent contamination with oxygen. At predetermined times, the cadmium concentration was determined by sampling the catholyte and analyzed it by atomic absorption spectrometry (Varian SpectrAA 200 spectrometer) using an air/acetylene flame. The performance of electrochemical system for removing cadmium was studied by data analysis via Taguchi method based on the same electrolysis time (40 min), while the electrolysis of each experiment was ended at 120 min.

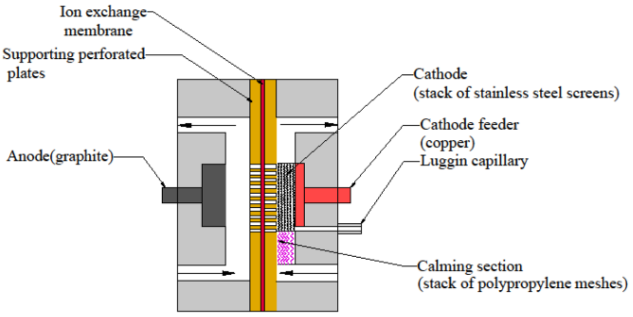

Fig. 2. The flow-by electrochemical cell.

Three stainless steel screens (316-AISI) were used with mesh numbers 30, 40, and 60 in^{-1}, respectively. Table 1 shows the properties of the screens. The porosity of screen was calculated by measuring weight/area density of the screen and using Eq. (1), then the specific surface area was calculated according to Eq. (2) [49]:

$$\varepsilon = 1 - \frac{m_s}{\rho_s l a_s} \tag{1}$$

$$s = (1 - \varepsilon) r \tag{2}$$

where ε is the porosity, r is the ratio of surface to volume of the wire (cm^{-1}) forming the screen, $r = 4/\text{d}$, m_s/a_s is the weight/area density (g/cm^2), l is the thickness of screen (cm), $l = 2$ d, ρ_s is the density of stainless steel 316-AISI (8.027 g/cm^3) [50], and s the specific surface area (cm^{-1}). The woven type of screen was determined by using Olympus BX51M with DP70 digital camera system and the diameter of wire (d) was measured by digital caliper.

The most important parameter in treatment of heavy metals is the applied current to the cell. Most of previous works did not explain the base on which they selected the range of current used in the experimental design [40,41]. In the present study, the current was selected to be equal to the limiting current under which the reaction rate is maximum. Therefore, linear voltagramms curves were proceeded at two levels: level-1 ([Cd] = 50 ppm, Q = 3 L/min, N = 30 in^{-1}) and level-2 ([Cd] = 200 ppm, Q = 7 L/min, N = 60 in^{-1}) as shown in Fig. 3. These voltagramms were obtained by increasing the current stepwise and measuring the steady-state cathode potential against a saturated calomel reference electrode. The limiting current is found at the middle point of a straight line

Table 1
Screen parameters

N (in^{-1})	30	40	60
Woven type	Plain square	Plain square	Twill
d (cm)	0.03	0.025	0.02
m_s/a_s (g/cm^2)	0.124	0.122	0.129
E	0.715	0.689	0.635
s (cm^{-1})	38.06	49.81	73.10

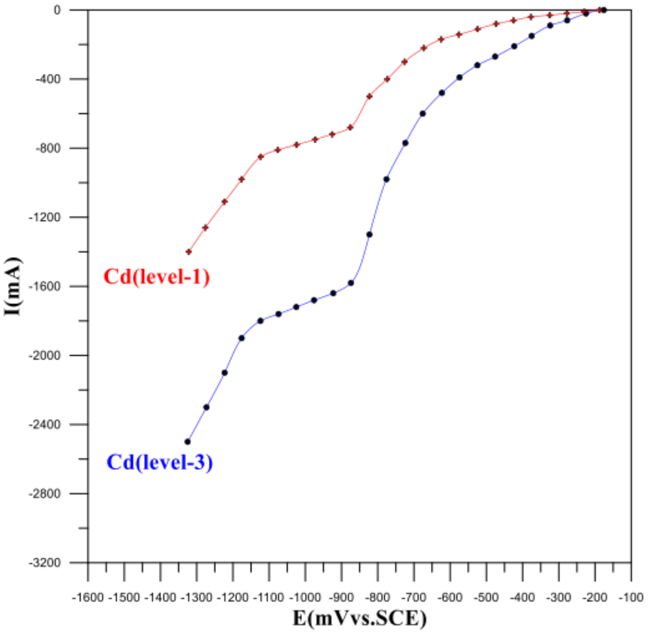

Fig. 3. Linear voltammetric curves for reduction of cadmium on the stack of stainless steel screens electrode at two levels: level-1 ([Cd] = 50 ppm, $Q = 3$ L/min, $N = 30$ in^{-1}), level-2 ([Cd] = 200 ppm, $Q = 7$ L/min, $N = 60$ in^{-1}). Supporting electrolyte (0.5 M NaCl + 0.1 M H_3BO_3), pH = 5, no. of screens = 5.

that follows the plateau region and is limited by E_{max} and E_{min} [51]. The limiting currents for reduction of cadmium at level-1 and level-2 were –0.78 and –1.72 A, respectively. The cathode potential related to these currents was –1025 mV vs. saturated calomel electrode (SCE).

2.2. Taguchi method

Conventional methods for experimental design are complex in nature and difficult to use. In addition, these methods also require a large number of experiments when the process parameters increases. In order to minimize the number of experiments, a powerful tool has been designed for high quality systems by Taguchi. Taguchi approach to design the experiments has got high adoptability and can be applied with confined knowledge of statistics, hence gained wide popularity in engineering application [52]. Four factors (concentration of cadmium, current, flow rate, and mesh number of screen) with three levels were selected as shown in Table 2. L_9 (3^4) standard orthogonal array presented in Table 3 was employed. This array is most suitable to provide the minimum degrees of freedom as 9 [= 1 + 4 × (3–1)] required for the experimental exploration. With the selection of L_9 (3^4) orthogonal array, the number of experiments required for each element is reduced to 9, which in classical combination method using full factorial experimentation would require $3^4 = 81$ number of experiments to find the influencing parameters [53]. To analyze the results, Taguchi method uses a statistical measurement of performance called S/N ratio, where signal represents the desirable value (i.e., the mean for the output characteristic) and noise represents the undesirable value (i.e., the square deviation for the output characteristic). Therefore, the S/N ratio is the ratio of mean square deviation. Its unit is dB. The S/N ratio equation depends on the criterion for the quality characteristic to be optimized. There are many different possible S/N ratios, however, three of them are considered standard and generally applicable in the most situations: larger is better (LTB), small is better (STB), and nominal is better (NTB). In this study, the LTB (Eq. (3)) is removal and current efficiencies while the STB (Eq. (4)) is the EC, the following relations are used for S/N calculations [53]:

$$\left(\frac{S}{N}\right)_{\text{LTB}} = -10\log\left[\frac{1}{n}\Sigma_{i=1}^{n}\frac{1}{y_i^2}\right] \tag{3}$$

$$\left(\frac{S}{N}\right)_{\text{STB}} = -10\log\left[\frac{1}{n}\Sigma_{i=1}^{n}y_i^2\right] \tag{4}$$

where n is the repetition number of each experiment under the same conditions for design parameters and y_i is the response of each experiment. A larger S/N noise ratio corresponds to better quality characteristics. Therefore, the optimal level of process parameters is the level of highest S/N ratio. The nine Taguchi experiments were conducted twice to ensure the reliability of experimental data for an S/N analysis. The performance was measured in terms of metal RE, CE, and EC [54].

The RE can be calculated as follows:

$$\text{RE} = \frac{C_i - C_o}{C_i} \times 100 \tag{5}$$

Table 2
Control factors and their levels in the experimental design

Levels	Factors			
	[Cd] (ppm)	I (A)	Q (L/min)	N (wire/in)
1	50	0.78	3	30
2	125	1.25	5	40
3	200	1.72	7	60

Table 3
Taguchi L_9 (3^4) orthogonal array design matrix

Exp. No.	Factors							
	Coded value				Real value			
	A	B	C	D	[Cd] (ppm)	I (A)	Q (L/min)	N (wire/in)
1	1	1	1	1	50	0.78	3	30
2	1	2	2	2	50	1.25	5	40
3	1	3	3	3	50	1.72	7	60
4	2	1	2	3	125	0.78	5	60
5	2	2	3	1	125	1.25	7	30
6	2	3	1	2	125	1.72	3	40
7	3	1	3	2	200	0.78	7	40
8	3	2	1	3	200	1.25	3	60
9	3	3	2	1	200	1.72	5	30

where RE is the removal efficiency (%); c_i is the initial metal concentration (ppm), c_o is the outlet metal concentration (ppm) after certain interval of time Δt (s). The CE is the yield based on the electric charge that reacted during electrolysis. It can be calculated as follows:

$$\mathrm{CE} = \frac{100 z_i \cdot F\Delta m}{M_i I \Delta t} \tag{6}$$

where CE is the current efficiency (%); z_i is the number of electrons; F is the Faraday constant (96,487 A s/mol); Δm (g) is the mass deposited at interval of time Δt (s); M_i is the molar mass (g/mol); and I is the applied current (A). The energy costs of the applied electrochemical process are closely related to the CE. The power consumption may be referred as the amount of substance on a molar, mass, or volume basis. The following relation was used to calculate it:

$$\mathrm{EC} = \frac{2.788 \times 10^{-4} VI\Delta t}{\Delta m} \tag{7}$$

where EC is the energy consumption (kWh/kg) and V is the cell potential (V).

3. Results and discussion

Table 4 shows the L_9 orthogonal array results considering two replicates per run. The analysis of these results is made with the help of software package MINITAB-17and using a general linear model.

3.1. Analysis of variance

Analysis of variance (ANOVA) is a statistical technique, which can infer some important conclusions based on analysis of the experimental data. The method is very useful for revealing the level of significance of influence of factor(s) on a particular response. It separates the total variability of the response into contributions of each of the factors and the error [53,55]. ANOVA table shows the sum of the square (Seq SS), the degree of freedom (DF), adjusted sum of squares (Adj SS), adjusted mean of square (Adj MS), the percentage contribution (Cr. %) of each parameter, *F* value, and *p* value. Percentage of contribution indicates the relative power of a factor to reduce variation. For a factor with a high percentage of contribution, a small variation will have a great influence on the performance [52]. Statistically, *F* test provides a decision at some confidence level as to whether these estimates are significantly different. Larger *F* value indicates that the variation of the process parameter makes a big change on the performance [56]. The *F* value for this condition with 95% confidence level is 19 [53]. *p* Value determine the significant of each factor on response where value of <0.05 (for a confidence level of 95%) indicates that the factor is significant. The results of ANOVA are summarized in Table 5.

It can be seen that current has the greatest effect on the RE with percentage of contribution (74.48%) while the other factors have no significant effect on the RE. On the other hand, concentration presents the higher effect on the CE and EC with a percentage of contribution 70.91% and 62.15%, respectively, followed by current. Also in this case, flow rate and mesh number have no significant effect on these responses. This behavior may be attributed to the higher reduction potential of cadmium on stainless steel electrode (–1,025 mV vs. SCE) which permits the side reaction (hydrogen evolution) to proceed at higher rate as the current increases or cadmium ion concentration decreases.

3.2. Analysis of signal-to-noise ratio

The S/N ratio in Taguchi method is one of the most important criteria for analysis of experimental data. Table 6 shows the S/N ratios calculated based on Eqs. (3) and (4) for all the responses of experiments presented in Table 4. The influence of each control factor on the responses is obtained from the response Table 7, which represented graphically in Fig. 4.

The response table shows the average of the selected characteristic for each level of the factors. It includes ranks based on delta statistics, which compare the relative magnitude of effects. The main effect plots are used to determine the optimal design conditions. Table 7 shows that current has rank 1 followed by mesh number, concentration, and flow rate with respect to RE, while concentration has rank 1

Table 4
Results of the experimental design

Exp. No.	Responses					
	RE (%)		CE (%)		EC (kWh/kg)	
	Replica (1)	Replica (2)	Replica (1)	Replica (2)	Replica (1)	Replica (2)
1	82.26	86.60	14.64	15.41	9.96	9.46
2	89.12	93.98	8.81	9.29	21.43	20.60
3	95.38	97.10	8.25	8.40	28.00	28.27
4	89.76	85.37	40.44	38.52	3.46	3.71
5	92.41	90.50	27.58	26.16	6.84	7.33
6	97.40	92.48	19.28	18.30	12.02	12.82
7	82.34	78.05	61.92	58.69	2.26	2.32
8	94.83	90.17	41.96	39.90	4.57	4.73
9	97.97	93.39	33.07	31.57	7.22	7.58

Table 5
Analysis of variance for cadmium removal

Source	DF	Seq SS	Cr. (%)	Adj SS	Adj MS	*F* value	*p* Value
Removal efficiency							
[Cd] (ppm)	2	16.66	2.92	16.66	8.33	0.86	0.46
I (A)	2	425.17	74.48	425.17	212.59	21.94	0.00
Q (L/min)	2	9.75	1.71	9.75	4.876	0.50	0.62
N (wire/in)	2	32.08	5.62	32.08	16.04	1.66	0.24
Error	9	87.20	15.28	87.20	9.69		
Total	17	570.86	100				
Model	*S*	R^2	Adj R^2	PRESS	Pred R^2		
Summary	3.11	84.72%	71.15%	348.81	38.90%		
Current efficiency							
[Cd] (ppm)	2	3,346.6	70.91	3,346.6	1,673.29	1,233.22	0.000
I (A)	2	1,101.5	23.34	1,101.5	550.74	405.89	0.000
Q (L/min)	2	156.07	3.31	156.07	78.03	57.51	0.000
N (wire/in)	2	103.11	2.18	103.11	51.55	37.99	0.000
Error	9	12.21	0.26	12.21	1.36		
Total	17	4719.45	100				
Model	*S*	R^2	Adj R^2	PRESS	Pred R^2		
Summary	1.17	99.74%	99.51%	48.85	98.96%		
Energy consumption							
[Cd] (ppm)	2	741.79	62.15	741.79	370.89	3,160.71	0.000
I (A)	2	349.68	29.30	349.68	174.84	1,489.96	0.000
Q (L/min)	2	38.39	3.22	38.39	19.19	163.56	0.000
N (wire/in)	2	62.58	5.24	62.58	31.29	266.65	0.000
Error	9	1.06	0.09	1.06	0.12		
Total	17	1,193.49	100				
Model	*S*	R^2	Adj R^2	PRESS	Pred R^2		
Summary	0.34	99.91%	99.83%	4.22	99.65%		

Table 6
The S/N ratios for all responses

Exp. No.	Responses		
	RE (%)	CE (%)	EC (kWh/kg)
	S/N ratio	S/N ratio	S/N ratio
1	38.52	23.53	–19.75
2	39.22	19.13	–26.45
3	39.67	18.41	–28.99
4	38.84	31.92	–11.10
5	39.36	28.58	–17.01
6	39.54	25.47	–21.87
7	38.07	35.60	–7.20
8	39.31	32.23	–13.35
9	39.61	29.91	–17.39

followed by current, flow rate, and mesh number at both CE and EC. It is clear that flow rate and mesh number have the lower and the same degree of effect on all responses. The results of response table are in agreement with ANOVA results. According to Fig. 4, the optimum levels of the controlling factors can be determined depending on the highest value of S/N ratio. The results are summarized in Table 8. It is clear that both CE and EC share the same optimum level of control factors.

Table 8 shows that two sets of optimum control factor levels can be adopted, the first based on RE while the second based on the CE and EC. Therefore, a comparison between these two optimum conditions must be done to determine the suitable one to be adopted. From the electrochemical engineering point of view, the performance of the electrochemical reactors is evaluated based on the possibility of achieving a higher RE as possible as with a suitable CE and a lower value of EC of the process [54]. Table 9 shows the predicted values of the three responses based on the two optimum levels of controlling factors. It was observed that the optimum level-1 gives higher RE with lower CE and higher EC while the optimum level-2 gives the opposite results.

By comparing optimum level-1 and conditions stated on experiment no. 9 with the neglecting the effect of mesh number, it is clear that starting from 200 ppm with RE higher than

Table 7
Response tables for signal-to-noise ratios for cadmium removal

	Level	[Cd] (ppm)	*I* (A)	*Q* (L/min)	*N* (in^{-1})
Removal efficiency (larger is better)	1	39.14	38.48	39.12	39.16
	2	39.24	39.30	39.22	38.95
	3	39.00	39.60	39.03	39.27
	Delta	0.25	1.13	0.19	0.33
	Rank	3	1	4	2
Current efficiency (larger is better)	1	20.35	30.35	27.08	27.34
	2	28.65	26.64	26.98	26.73
	3	32.58	24.59	27.53	27.52
	Delta	12.23	5.75	0.54	0.79
	Rank	1	2	4	3
Energy consumption (small is better)	1	–25.06	–12.68	–18.33	–18.05
	2	–16.67	–18.94	–18.31	–18.51
	3	–12.65	–22.75	–17.73	–17.81
	Delta	12.42	10.07	0.60	0.70
	Rank	1	2	4	3

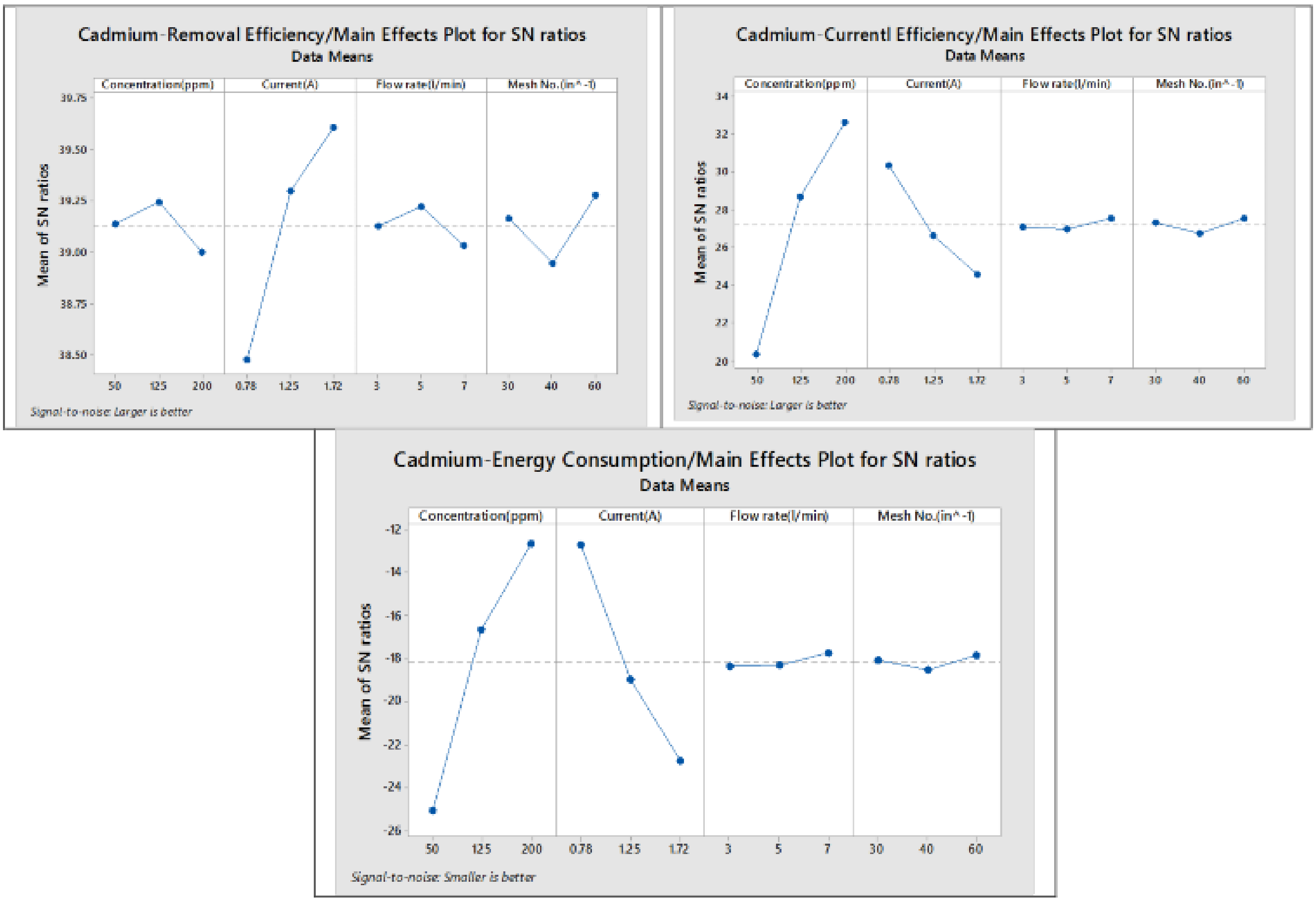

Fig. 4. Main effect plots of S/N ratios for copper removal.

96% and CE higher than 30% is better than the starting with 125 ppm because more cadmium ions can be removed with a lower EC. A RE of 99% with a CE of 28% were obtained with a level of EC 9 kWh/kg when the time of operation at experiment no. 9 was extended to 50 min. In the light of this comparison, adopting the conditions of experiment no. 9 with electrolysis time extended to 50 min will be more economic and compatible with legislations of environment where cadmium should be discharged at concentration lower than 2 ppm.

3.3. Comparison with the previous studies

The comparison of experimental data with the previous works was performed by analyzing the removal and current efficiencies for the electrochemical systems used fixed bed electrodes. Table 10 shows the comparison of the present work with previous works. The current density in the present work was calculated based on the volume and specific surface of the electrode.

The results indicate that the present system is able to reduce concentration of cadmium from 200 to lower than 2 ppm through 50 min which is lower than the half value of time that required in the work of de Lourdes Llovera-Hernández et al. [57]. This is an indication that the present flow-by configuration is more efficient than flow-through configuration. In comparison with Dutra et al. [58] results, the time is reduced to 60% in the present work which refers to the high performance of the present work to remove of cadmium in spite of the higher CE observed in Dutra work due to the potentiostatic operation. The higher performance of present work may be attributed to the high turbulence-promoting ability, high porosity, and relatively low pressure drop that the present configuration was pertained. However, the present system somewhat results in a higher EC in comparison with the others because the current density used is double that used in the previous works consequently a higher voltage drop is occurred leading to more EC.

Table 8
Optimum levels of the control factors

Responses	Factors			
	[Cd] (ppm)	*I* (A)	*Q* (L/min)	*N* (in^{-1})
RE (%)	125	1.72	5	60
CE (%)	200	0.78	7	60
EC (kWh/kg)	200	0.78	7	60

4. Conclusions

Taguchi methodology has been successfully applied to find out the parameters of cadmium removal process, which maximized the CE and minimized the EC. Taguchi analysis has suggested that among the various parameters, concentration and current has the largest effect on the performance of electrochemical removal of cadmium. Two sets of optimum control factors levels were obtained via analysis of Taguchi design, the first based on optimization of RE, while the second based on optimization of CE and EC. However, no one of these levels was adopted because one of Taguchi experiments (no. 9) gave better result when the time of operation extended to 50 min where a higher CE and a lower EC at the same RE of the optimum level-1 were obtained. As a final conclusion the following operating conditions may be considered the optimum for removal of cadmium [Cd] = 200 ppm, *I* = 1.72 A, *Q* = 5 L/min, *N* = 30 in^{-1}, and electrolysis time 50 min. Based on these operating conditions a RE of 99% was achieved

Table 9
The predicted values of current efficiency for two optimum levels of control factors

Optimum level	Control factor				Predicted removal efficiency (%)		Predicted current efficiency (%)		Predicted energy consumption (kWh/kg)	
	[Cd] (ppm)	*I* (A)	*Q* (L/min)	*N* (in^{-1})	Fit	95% CI	Fit	95% CI	Fit	95% CI
1	125	1.72	5	60	99.11	(94.13; 104.09)	20.69	(18.82; 22.55)	14.38	(13.66; 15.1)
2	200	0.78	7	60	83.41	(78.43; 88.39)	60.50	(58.63; 62.36)	2.51	(2.38; 2.63)
Exp. 9	200	1.72	5	30	96.1		32.32		7.4	

Table 10
Comparison with previous works

Parameters	Author		
	Present work	de Lourdes Llovera-Hernández et al. [57]	Dutra et al. [58]
Operation mode	Galvanostatic	Galvanostatic	Potentiostatic
Cathode type	Flow-by stainless steel screen	Flow-through RVC	Flow-by RVC
Initial concentration (ppm)	200	225	200
pH	5	7	2.0
Current density (C.D) (A/m^2)	7.5	3.5	$E = -0.89$ V (SCE)
Time (min)	50	120	85
RE (%)	99.0	99.0	99.8
CE (%)	28	34	40

and an EC of 9 kWh/kg was required. Due to the high turbulence-promoting ability of the present system, it could be used for removing other heavy metals like zinc, lead, and copper with more efficient.

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