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Taguchi Modeling for Techno-Economical Evaluation of Cr^{+6} Removal by Electrocoagulation Process With the Aid of Two Coagulants

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ABSTRACT: The research aimed to apply the Taguchi method for techno-economical evaluation of Cr^{+6} removal using the electro-coagulation process with the aid of two different coagulants ($FeCl_3$ and PAC). Taguchi orthogonal array L27 (3^{13}) was applied for analyzing the effect of four variables including initial pH, reaction time, current density and coagulant types in an attempt to improve the chromium removal efficiency. Based on the signal-to-noise ratio (S/N) and the analysis of variance (ANOVA), the results indicated that the reaction time was the most important variable on the hexavalent chromium removal efficiency. However, the current density, reaction time and coagulant types significantly influenced the operating costs. The optimum conditions for the mentioned variables were found to be an initial pH of 7, a reaction time of 60 min, a current density of 12.5 mA/cm^2 and $FeCl_3$ as a coagulant. Due to the interaction between the initial pH and coagulant type at pH 7, PAC also considered as a coagulant in this experiment. Under the mentioned conditions, the removal efficiencies of 92% and 95% were achieved using the chromium removal process by $FeCl_3$ and PAC, respectively.

Keywords: Chromium Removal Efficiency, Electrocoagulation Process, Operating Costs, Taguchi Design.

INTRODUCTION

Wastewater containing chromium is discharged into the environment by different industries such as leather tanning, metal plating, textile dyeing, production of fertilizers and chemistry (Aber et al., 2009, Zongo et al., 2009). Chromium generally exists in aqueous solution with two stable oxidation states: hexavalent (Cr^{+6}) and trivalent (Cr^{+3}). Hexavalent chromium is known to be toxic and carcinogenic, causing health problems such as vomiting, liver damage and pulmonary congestions, whereas trivalent chromium is less toxic and can be precipitated out of solution in the form of $Cr(OH)_3$ (Eary and Dhanpat, 1988, Yurik and Pikaev, 1999).

The electro-coagulation (EC) technique has attracted a great deal of attention because of the inherent simplicity of its design and operation. In addition, the EC effluent has good quality before its discharge into the aquatic environment (Secula et al., 2012). The electrolytic reactions during the EC process with Al electrodes are

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summarized inhere, as follows (Chafi et al., 2011; Desal, 2011):

Anodic reaction:

$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^{-} \quad (1)$$

Cathodic reaction:

$$2H_2O + 2e^{-} \rightarrow 2OH^{-}_{(aq)} + H_{2(g)} \quad (2)$$

Chromium removal using the electrocoagulation technique has been applied by many investigators (Akbal and Camci, 2010, 2011; Asadi Habib et al., 2012; Bazrafshan et al., 2008; Bhatti et al., 2011; Heidmann and Calmano, 2008; Keshmirzadeh et al., 2011). The electrocoagulation process using Al electrodes does not seem to be very efficient for the removal of Cr^{+6} from wastewater (Barrera-D´ıaz, 2012; Bhatti et al., 2011). The use of coagulant such as $FeCl_3$ and polyaluminum chloride (PAC) in conjunction with the EC process to improve the Cr^{+6} removal efficiency has been rarely considered.

Taguchi is the preferable method among statistical experimental design methods (Keles, 2009) since it uses a special design of an orthogonal array (OA) to obtain the best variables for the optimum process design with the least number of runs (Irdemez et al., 2006; Martínez-Villafa˜ne and Montero-Ocampo, 2010). A wide range of applications using the Taguchi method has been seen for the treatment of various pollutants (Asadi Habib et al., 2012; Davila et al., 2011; Irdemez et al., 2006; Martínez-Villafa˜ne and Montero-Ocampo, 2010 andYidiz, 2008). However, in recent researches, the Taguchi method has been rarely used for the optimization process with two responses, including, for instance, Cr^{+6} removal and its operation costs. In addition to that, the techno-economical evaluation in the electro-coagulation process using different types of experimental design methods has been considered by other researches in this context (Bhatti et al., 2009 and Bhatti et al., 2011).

This research aims to apply the Taguchi method for the techno-economical evaluation of Cr^{+6} removal using the electro-coagulation process with the aid of two different coagulants (i.e. $FeCl_3$ and PAC). To do this, the relationship between two responses (i.e. removal efficiency and operating costs) and four variables including initial pH, reaction time, current density and coagulant types is determined by OA L27 (3^{13}) at three levels.

MATERIALS AND METHODS

EC Apparatus

A plexiglas tank with dimensions of 13 cm × 15 cm × 18 cm and an effective volume of 2.5 lit was employed for the present study. Four Al plate electrodes (Arak Co. (Iran), 99% purity) with a total effective area of 240 cm^2 were used. Thickness of aluminum plates was considered to be 3 mm and inter-electrodes distance was maintained at 2 cm in each experiment. The electrodes were connected to a DC power supply (Micro, PW4053R, 0-5 A, 0-40 V) in a monopolar mode.

Experimental Procedure and Calculation of Operating Costs

Chromium solutions (50 mg/l) were prepared by dissolving the required amount of potassium dichromate (Merck, 99.9% purity) in distilled water. Sodium chloride salt (Merck) was used to adjust the initial solution conductivities (2.5 mS/cm). PAC (30% w/w Al_2O_3) in powder and $FeCl_3.6H_2O$ (97% purity) in powder (each 750 mg/l) were added to the electrocoagulation tank, and the pH values were adjusted by NaOH and H_2SO_4 (4 N)

(Merck), and measured using the pH meter (340i, WTW, Germany). All experiments were accomplished at room temperature.

The analysis of chromium was carried out using the UV-visible spectrophotometer (HACH, DR 4000, USA) adopted from the standard methods (3500- Cr B) for examination of water and wastewater (APHA, 1998).

In the electro-coagulation process, operating costs consist of costs of chemicals, materials of electrodes, electrical energy and final disposal of sludge (Behbahani et al., 2011). In this research, an economic evaluation was undertaken for energy, electrode material and coagulant costs in the following form:

Operating cost = a.C energy + b.C electrode + c.C coagulant

where operating cost (US$/m^3), C energy (kWh/m^3), C electrode (kg Al/m^3) and C coagulant (kg PAC or FeCl$_3$/m^3) are consumption quantities for the treatment of chromium solutions. Coefficients a, b and c are the Iranian market prices in 2012 as follows:

Coefficient a: industrial electrical energy price=0.0352 US$/kWh

Coefficient b: wholesale electrode material price=5.33 US$/kg Al.

Coefficient c: wholesale PAC price=0.98 US$/kg PAC and wholesale FeCl$_3$ price=2.21 US$/kg FeCl$_3$.

The energy consumption in the EC process at a constant voltage and current was calculated by Eq. (3):

$$E = \frac{U.I.t}{V} \tag{3}$$

where E is the energy consumption (kWh/m^3), U is the applied voltage (V), I is the current intensity (A), t is the electrocoagulation time (h), and V is the volume of wastewater (lit).

Mass depletion of the electrodes was calculated by subtracting the weights of the electrodes at the end of each experiment from their weights at the beginning of a corresponding experiment.

Taguchi Design

The variables selected for this investigation are initial pH, reaction time, current density and coagulant types. Table 1 shows the selected variables and their levels in the Taguchi design. Initial chromium concentration was kept in 50 mg/l for the statistical analysis purpose.

In the Taguchi method, if all variable interactions, are taken into consideration, the L27 (3^{13}) orthogonal array is the most appropriate experiment plan. The L27 orthogonal array with variables and their interactions are shown in Table 2. As seen in Table 2, the first, second, fifth, and ninth columns are respectively assigned to initial pH (A), reaction time (B), current density (C), and coagulant types (D). The remaining columns are assigned to their interactions.

Table 1. Experimental range and levels of independent parameters in Taguchi design.

Variables	Symbol	Level		
		1	2	3
Initial pH	A	3	7	10
Reaction time (min)	B	20	40	60
Current density (mA/cm^2)	C	4.16	8.33	12.5
Coagulant types (mg/l)	D	PAC	FeCl$_3$	EC

Table 2. L27 Orthogonal array with factors and their interactions.

Trial No.	Column No.												
	1	2	3	4	5	6	7	8	9	10	11	12	13
	A	B	(A×B), (C×D)	(A×B)	C	(A×C) ,(B×D)	(A×C)	(B×C) ,(A×D)	D	(A×D)	(B×C)	(B×D)	(C×D)
1	1	1	1	1	1	2	2	1	1	1	1	1	1
2	1	1	1	1	2	3	3	2	2	2	2	2	2
3	1	1	1	1	3	2	3	1	3	3	3	3	3
4	1	2	2	2	1	3	1	2	2	2	3	3	3
5	1	2	2	2	2	1	2	3	3	3	1	1	1
6	1	2	2	2	3	2	3	2	1	1	2	2	2
7	1	3	3	3	1	3	1	3	3	3	2	2	2
8	1	3	3	3	2	1	2	1	1	1	3	3	3
9	1	3	3	3	3	2	3	3	2	2	1	1	1
10	2	1	2	3	1	3	1	1	2	3	1	2	3
11	2	1	2	3	2	1	2	2	3	1	2	3	1
12	2	1	2	3	3	3	2	1	1	2	3	1	2
13	2	2	3	1	1	1	3	2	3	1	3	1	2
14	2	2	3	1	2	2	1	3	1	2	1	2	3
15	2	2	3	1	3	3	2	2	2	3	2	3	1
16	2	3	1	2	1	1	3	3	1	2	2	3	1
17	2	3	1	2	2	2	1	1	2	3	3	1	2
18	2	3	1	2	3	3	2	3	3	1	1	2	3
19	3	1	3	2	1	1	3	1	3	2	1	3	2
20	3	1	3	2	2	2	1	2	1	3	2	1	3
21	3	1	3	2	3	2	2	1	2	1	3	2	1
22	3	2	1	3	1	3	3	2	1	3	3	2	1
23	3	2	1	3	2	2	3	1	2	1	1	3	2
24	3	2	1	3	3	3	1	2	3	2	2	1	3
25	3	3	2	1	1	1	2	3	2	1	2	1	3
26	3	3	2	1	2	2	3	2	3	2	3	2	1
27	3	3	2	1	3	3	1	3	1	3	1	3	2

Experimental variables, their levels, and the results of conducted experiments on responses of the Cr^{+6} removal efficiency and operating costs are presented in Table 3. The model terms were evaluated by the F-value with a 95% confidence level.

Table 3. Observed results of experiments corresponding to Taguchi design.

Experimental No.	Variables and their levels				Chromium removal (%)	Operating Cost (US$/m^3)
	A	B	C	D		
1	3	20	4.16	PACl	11.6	1.075
2	3	20	8.33	Fecl$_3$	63.0	2.750
3	3	20	12.5	EC	63.8	1.520
4	3	40	4.16	Fecl$_3$	70.0	2.570
5	3	40	8.33	EC	75.8	2.070
6	3	40	12.5	PACl	64.4	2.770
7	3	60	4.16	EC	69.8	1.720
8	3	60	8.33	PACl	50.0	2.720
9	3	60	12.5	Fecl$_3$	80.6	7.065
10	7	20	4.16	Fecl$_3$	44.4	2.210
11	7	20	8.33	EC	27.6	0.824
12	7	20	12.5	PACl	57.4	1.730
13	7	40	4.16	EC	39.0	1.420
14	7	40	8.33	PACl	87.4	2.500
15	7	40	12.5	Fecl$_3$	68.6	4.880
16	7	60	4.16	PACl	67.6	1.920
17	7	60	8.33	Fecl$_3$	89.0	4.990
18	7	60	12.5	EC	85.0	5.700
19	10	20	4.16	EC	2.2	0.863
20	10	20	8.33	PACl	13.0	1.260
21	10	20	12.5	Fecl$_3$	41.4	3.370
22	10	40	4.16	PACl	20.6	2.000
23	10	40	8.33	Fecl$_3$	37.8	3.560
24	10	40	12.5	EC	59.0	3.000
25	10	60	4.16	Fecl$_3$	40.4	3.480
26	10	60	8.33	EC	59.6	3.100
27	10	60	12.5	PACl	70.0	4.360

The Taguchi method recommends the use of the loss function to measure the performance characteristics deviating from the desired value (Roy, 1990). The value of the loss function is further transformed into a signal-to-noise (S/N) ratio. In this study, the experimental observations were transformed into a signal-to-noise (S/N) ratio. Two categories of S/N ratios were used in this investigation: the larger the better for the Cr^{+6} removal efficiencies and the smaller the better for the operating cost. Two performance characteristics were evaluated by Eq. (4), for the larger the better and Eq. (5), for the smaller the better (Phadke, 1989):

$$\frac{S}{N} = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right) \tag{4}$$

$$\frac{S}{N} = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}y_i^2\right) \tag{5}$$

where S/N is the performance characteristics, n is the number of replicates and y_i is the response variable at (i = n).

RESULTS AND DISCUSSION

The analysis of variance (ANOVA) and S/N ratio were evaluated to determine the effect of each selected variable on the optimization criteria. For these analyses, the Minitab14 software was utilized. The results of the ANOVA for the Cr^{+6} removal efficiency and operating costs are shown Tables 4 and 5, respectively. In general, the F values and contribution (ρ) ratios indicate the importance of these variables in the electro-coagulation process.

Table 4. ANOVA for the response of Cr^{+6} removal efficiency (%) in Taguchi method.

Source	DF	Seq SS	Adj SS	Adj MS	F	P	ρ
A	2	238.27	238.27	119.13	6.89	0.028	18.75
B	2	354.54	354.54	177.27	10.25	0.012	27.90
C	2	233.86	233.86	116.93	6.76	0.029	18.40
D	2	58.72	58.72	29.36	1.70	0.260	4.62
A*B	4	88.77	88.77	22.19	1.28	0.373	6.98
A*C	4	87.54	87.54	21.89	1.27	0.379	6.89
A*D	4	105.12	105.12	26.28	1.52	0.308	8.27
Error	6	103.74	103.74	17.29			8.16
Total	26	1270.57					100.00

ρ = contribution ratio

Table 5. ANOVA for the response of operating costs (US$/$m^3$) in Taguchi method.

Source	DF	Seq SS	Adj SS	Adj MS	F	P	ρ
A	2	1.147	1.147	0.573	0.11	0.897	0.19
B	2	234.640	234.640	117.320	22.55	0.002	39.13
C	2	147.037	147.037	73.518	14.13	0.005	24.50
D	2	172.169	172.169	86.085	16.55	0.004	28.68
A*B	4	5.536	5.536	1.384	0.27	0.890	0.92
A*C	4	5.355	5.355	1.339	0.26	0.895	0.89
A*D	4	3.106	3.106	0.776	0.15	0.957	0.52
Error	6	31.210	31.210	5.202			5.2
Total	26	600.200					100.00

ρ = contribution ratio

As presented in Table 4, initial pH, reaction time, current density, and contribution ratios associated with coagulant types are 18.75%, 27.90%, 18.40% and 4.62% for the selected chromium removal efficiency, respectively. In addition, Table 5 shows the contribution ratios of 0.19%, 39.13%, 24.50% and 28.68% for initial pH, reaction time, current density, and coagulant type, respectively. Therefore, the reaction time is the most effective variable in the Cr^{+6} removal efficiency and its operating costs using the electro-coagulation process. On the other hand, according to the data presented in Table 4, the coagulant type and the selected interactions are the least significant variables in Cr^{+6} removal efficiency. Also, as shown in Table 5, all independent variables and the selected interactions are insignificant in the operating costs except the reaction time (39.13%), current density (24.50%), and coagulant type (28.68%).

The main effects of independent variables (A, B, C and D) on the responses of the Cr^{+6} removal efficiency and operating costs, using Taguchi method, are shown in Figures 1 and 2, respectively. Figures 1 and 2 illustrate the average of S/N ratios for each variable at the three different levels and the corresponding response variables.

According to Figure 1, by increasing the reaction time and current density, the Cr^{+6} removal efficiency will be improved. It is seen that the maximum removal efficiency was obtained when the initial pH of solutions was 7. Also, the results indicated that $FeCl_3$ as a coagulant outperforms PAC with regard to the chromium removal efficiency.

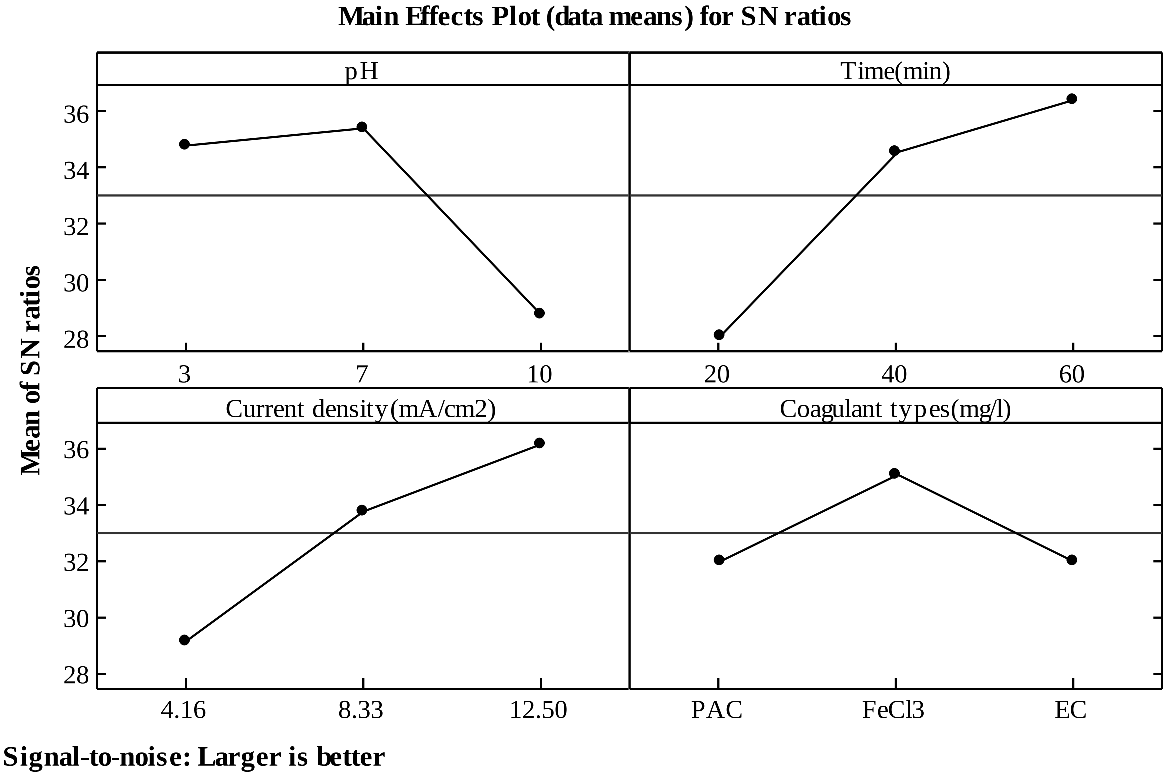


Fig. 1. Main effects plot of the independent parameters for the response of Cr^{+6} removal efficiency (%) in Taguchi method.

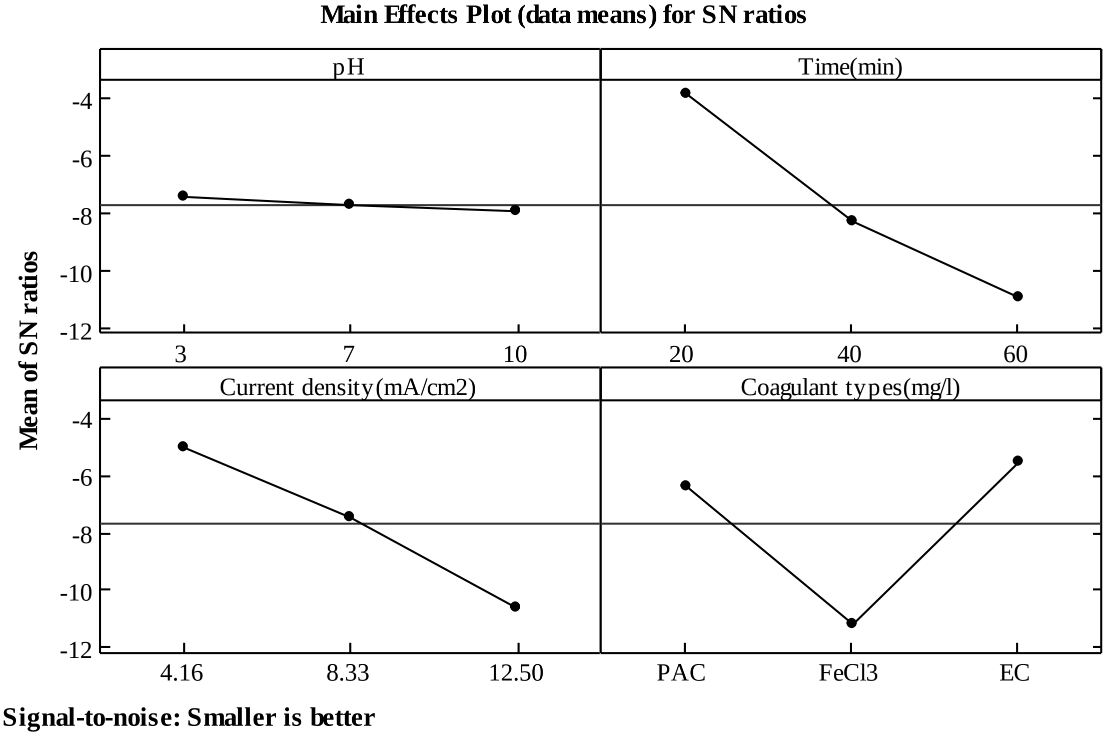


Fig. 2. Main effects plot of the independent parameters for the response of operating costs (US$/m^3) in Taguchi method.

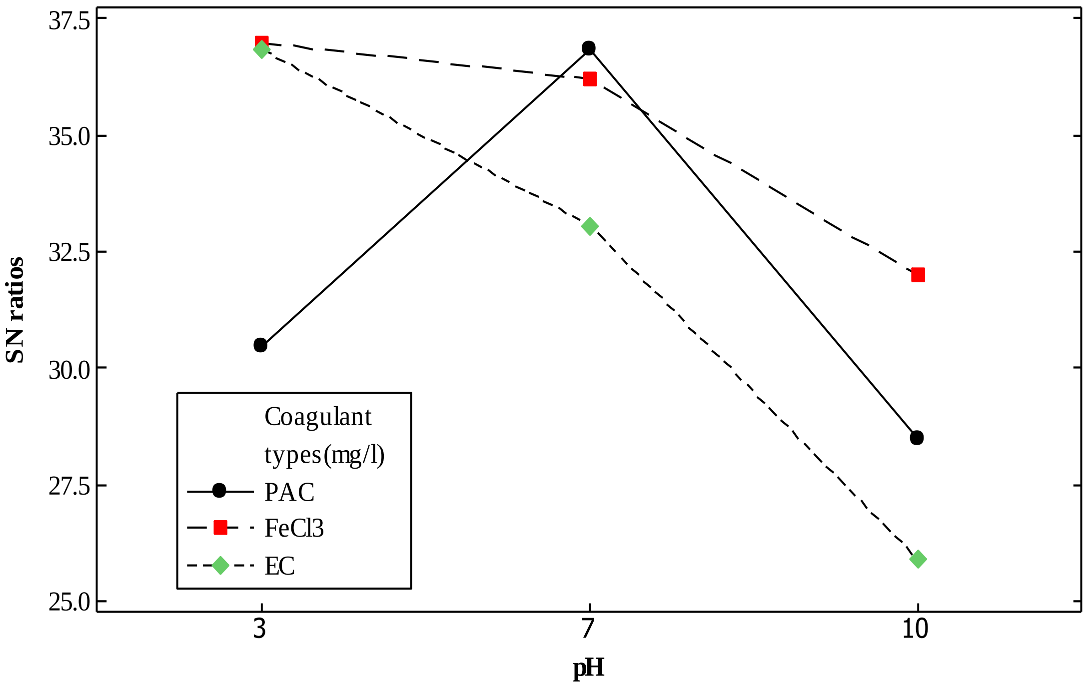
In the case of the operating costs, as illustrated in Figure 2, the operating costs are highly dependent on the reaction time, current density, and coagulant type. The amount of energy consumption (according to Eq. (3)) and electrodes corrosion (according to Faraday's law, Eq. (6)) increases as the current densities and reaction time go up.

$$m = \frac{\mathrm{ItM}}{\mathrm{zF}} \tag{6}$$

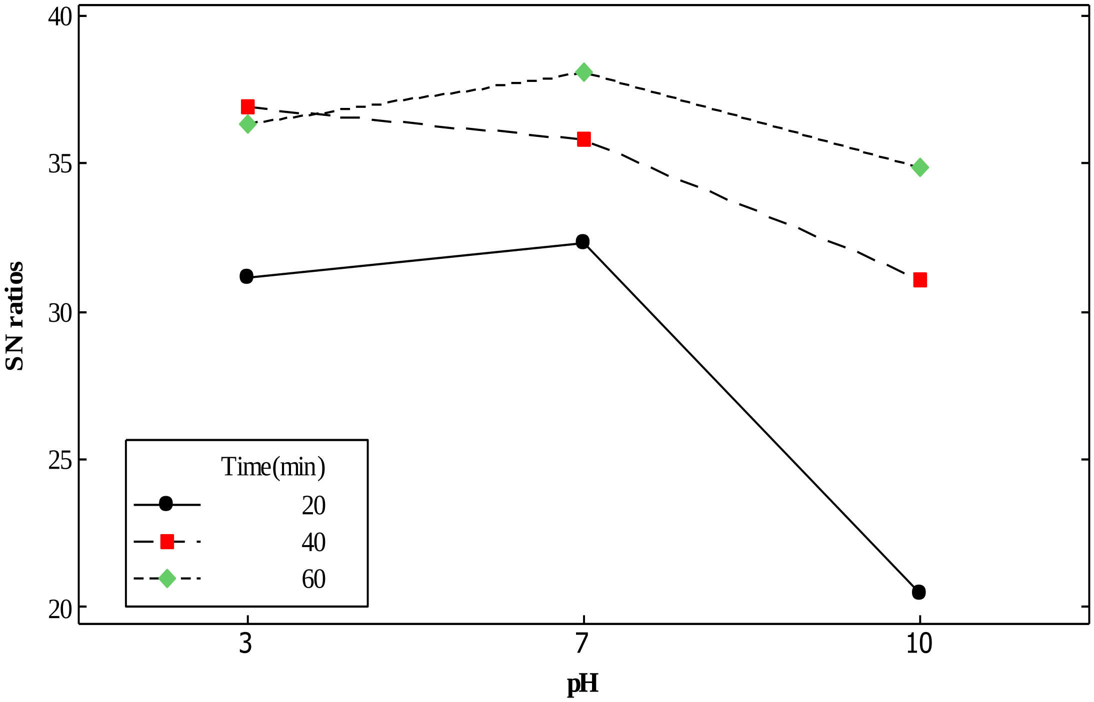
where I is the current (A), t is the time, M is the molecular weight of aluminum ion (g/mol), z is the number of electrons transferred in the reaction (3 for aluminum) and F is Faraday's constant (96486 C/mol) (Shafaei et al., 2011). Increasing the values of energy consumption, electrodes corrosion and coagulant dosages leads to higher operating costs. As depicted in Figure 2, the operating costs also increase by selecting $FeCl_3$ as a coagulant. Additionally, the findings revealed that the initial pH has no significant effect on the operating costs.

The interactions plot for the Cr^{+6} removal efficiency and operating costs are presented in Figure 3. As demonstrated in this Figure, the most important interaction is associated with coagulant type and initial pH for the Cr^{+6} removal efficiencies and operating costs.

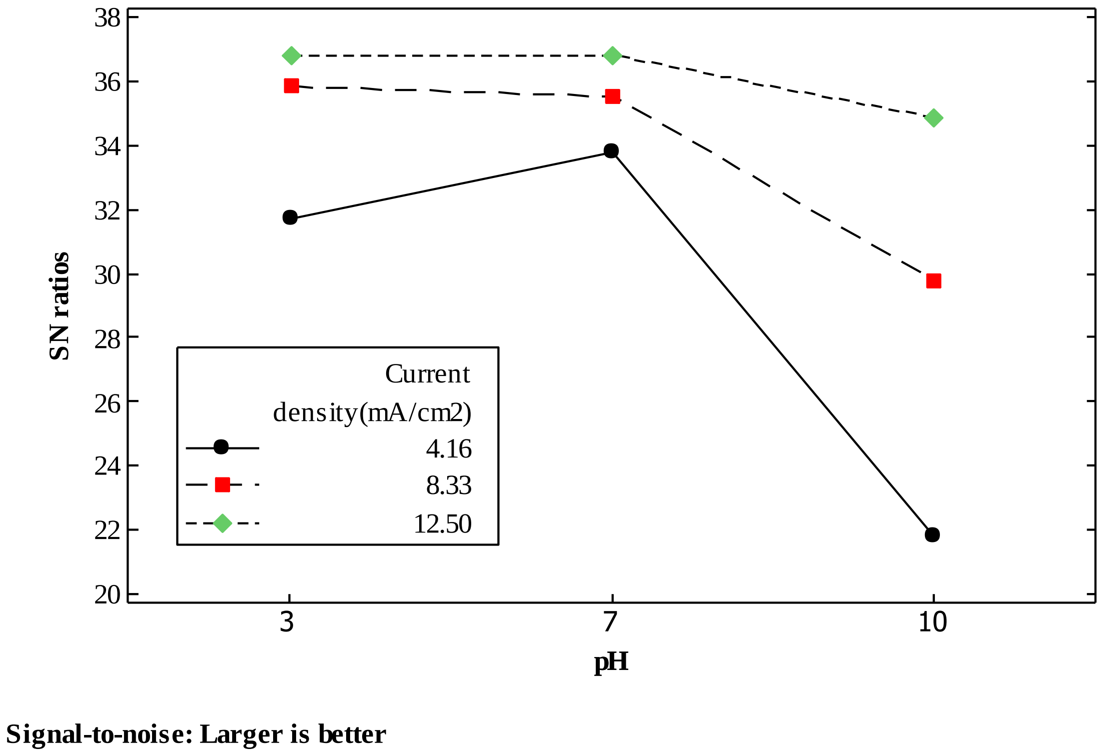
As seen in Figure 3, the interaction between the initial pH and coagulant type at pH 7 caused that the Cr^{+6}removal efficiency with the aid of PAC becomes higher than the Cr^{+6} removal efficiency with the aid of $FeCl_3$. Furthermore, the chromium removal process using PAC is cheaper than the chromium removal process using $FeCl_3$ at pH 7.



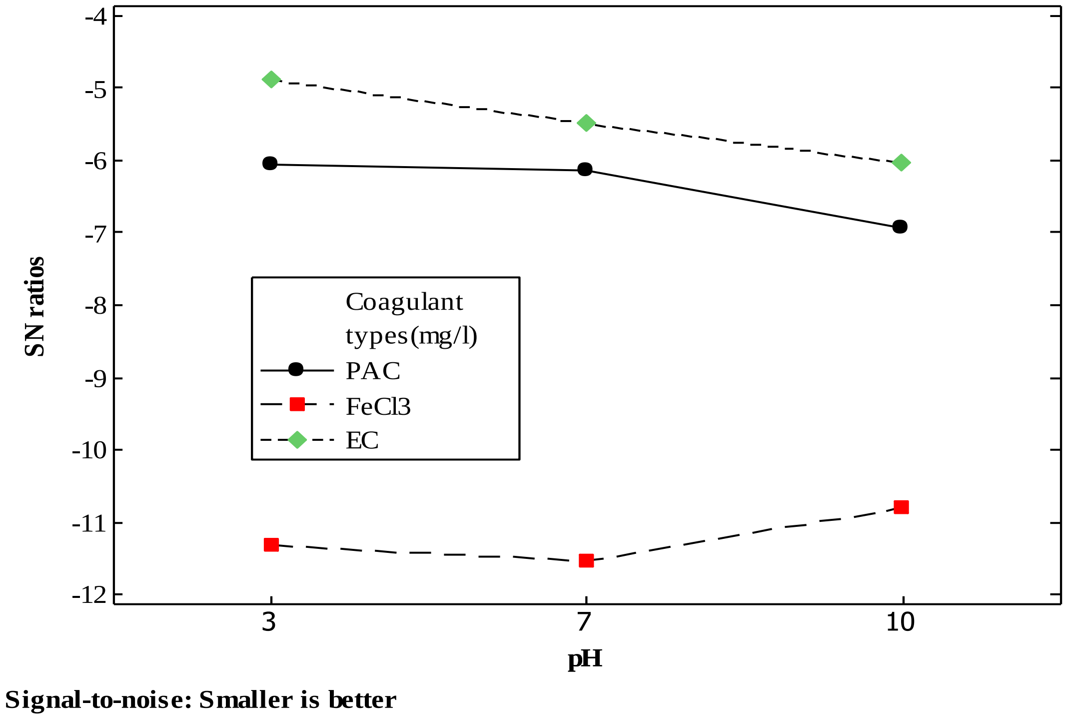
Signal-to-noise: Larger is better

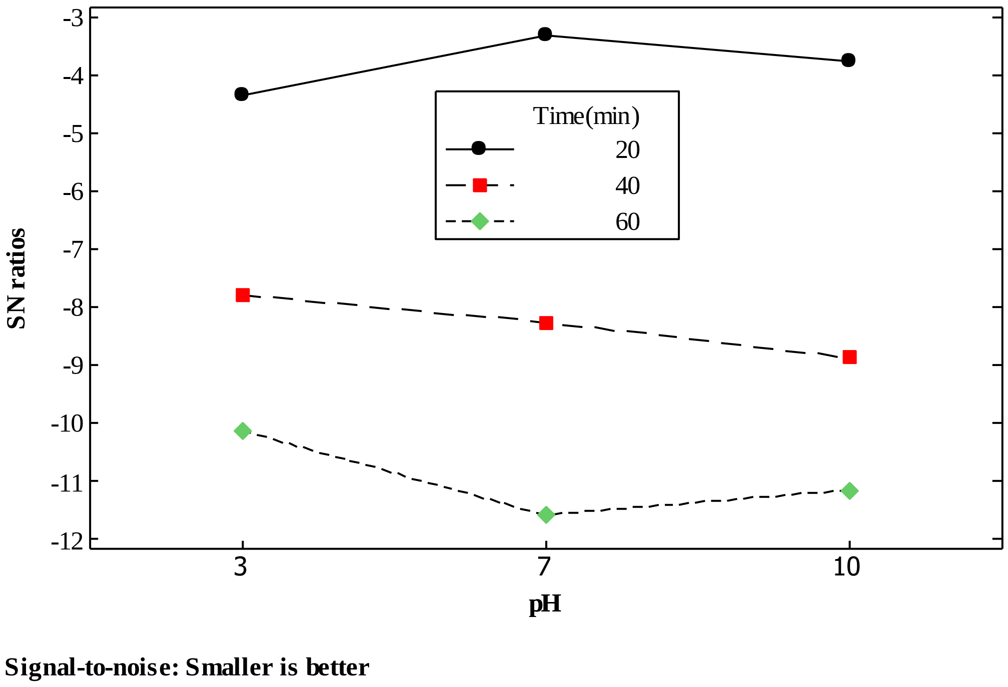


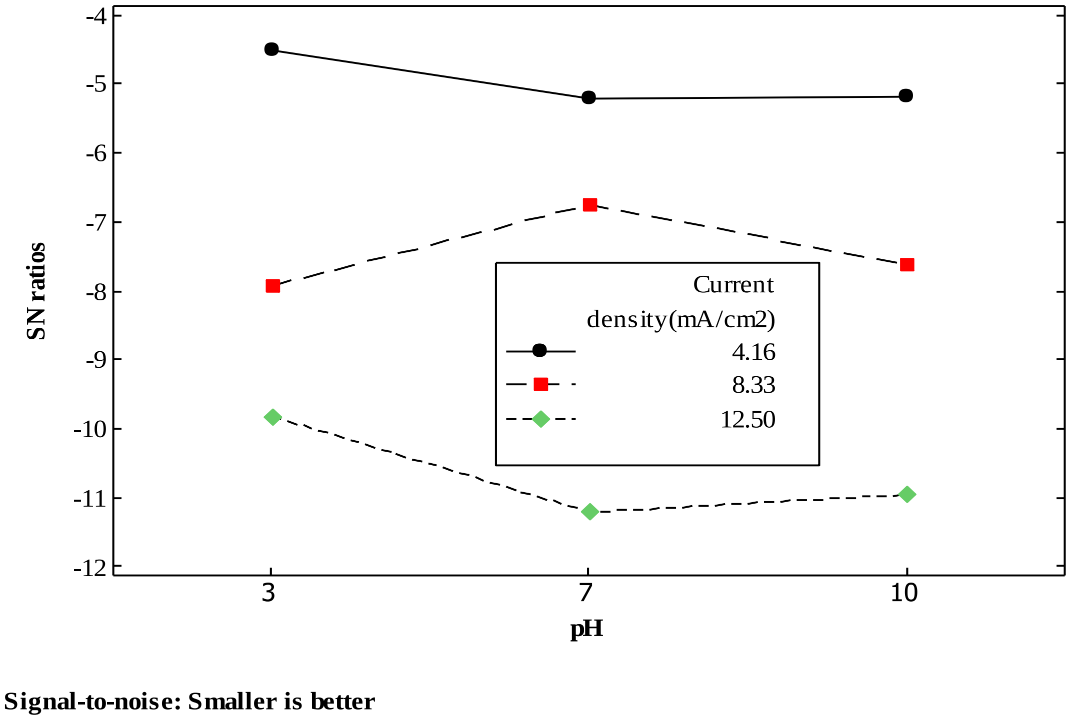
Signal-to-noise: Larger is better


38
36
34
32
30
28
26
24
22
20
SN ratios
3
7
10
pH
Current density(mA/cm2)
4.16
8.33
12.50
Signal-to-noise: Larger is better

(a)


-4
-5
-6
-7
-8
-9
-10
-11
-12
SN ratios
3
7
10
pH
Coagulant types(mg/l)
PAC
FeCl3
EC
Signal-to-noise: Smaller is better





(b)

Fig. 3. Interactions plot for the responses of: (a) Cr^{+6} removal efficiency (%) and (b) operating costs (US$/$m^3$) in Taguchi method.

Considering the maximum value of the S/N ratio as the optimum, the corresponding variables are as follows: an initial pH of 7, areaction time of 60 min, a current density of 12.5 mA/cm^2 and $FeCl_3$ as a coagulant. However, according to the interaction plot, there are interactions between the coagulant type and initial pH for the Cr^{+6} removal efficiency. Therefore, the optimum condition needs to be modified and the performance at the new optimum condition (initial pH of 7, reaction time of 60 min, current density of 12.5 mA/cm^2 and PAC as a coagulant) should be compared with the original optimum condition. Under the mentioned conditions, the response obtained from the conformation tests for the Cr^{+6} removal efficiency and operation cost were 92% and 6.5 US$/m^3 for the EC process using $FeCl_3$, and 95% and 4.75 US$/m^3 for the EC process using PAC, respectively. It can be said that the latter is associated with the higher removal efficiency and lower operating cost. So, it is confirmed as the optimum condition.

CONCLUSIONS

In this study, an orthogonal Taguchi L 27(3^{13}) array was employed for the techno-economical process optimization of hexavalent chromium removal using the electro-coagulation process. For this purpose, four variables including initial pH, reaction time, current density and coagulant types at three levels were examined. According to the ANOVA results, the reaction time was the most significant variable for the Cr^{+6} removal efficiency and operating costs. However, the coagulant type and the selected interactions were the least significant variables in the chromium removal efficiency. All independent variables and the selected interactions were also insignificant in operating costs except the reaction time (39.13%), current density (24.50%), and coagulant type (28.68%).

The optimum conditions for initial pH, reaction time, current density and coagulant types were equal to 7, 60 min, 12.5 mA/cm^2 and $FeCl_3$, respectively. Under these conditions, the removal efficiency of 92% and operation costs of 6.5 US$/m^3 were achieved. However, the interaction between the coagulant type and initial pH at pH 7 led to consider PAC as a selected coagulant. In the optimum conditions using PAC, the removal efficiency of 95% and operation costs of 4.75 US$/m^3 were reached. Thus, PAC was considered as a better option than $FeCl_3$ at pH 7 in the electro-coagulation process.

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