

57 (2016) 2389–2399 February



Optimization of operational parameters of electrocoagulation process for real textile wastewater treatment using Taguchi experimental design method

Fuat Ozyonar

Department of Environmental Engineering, Cumhuriyet University, 58140 Sivas, Turkey, Tel. +90 346 219 10 10, ext. 1582; Fax: +90 346 219 11 77; email: fozyonar@cumhuriyet.edu.tr

Received 15 August 2014; Accepted 4 January 2015

ABSTRACT

In this paper, Taguchi method was applied to determine the optimum operating conditions for textile wastewater treatment by electrocoagulation (EC) with iron electrodes. The removal efficiency of chemical oxygen demand (COD) and turbidity was considered as the performance parameters. L_9 orthogonal array (OA, three factors in three levels) was selected as the experimental design for three controllable parameters mentioned above. These parameters that mainly affect the EC process were specified as initial pH, current density and electrolysis time. Each parameter was examined at three levels using Taguchi method. Performance analysis was followed by a variance analysis in order to determine the optimum levels and relative magnitude of the effect of parameters. Taguchi's "the larger the better" performance formula was used. The optimum operating conditions in terms of COD and turbidity were found to be initial pH of 7 and 8, current density of 150 A/m^2 and electrolysis time of 20 and 30 min. At the optimum conditions, COD and turbidity removal efficiencies were obtained as 42.2 and 99.1%, respectively. Operating costs of EC process for COD and turbidity removals were calculated as 1.52 and $1.73 \notin m^3$. Besides, the UV-Vis spectrum was used to detect the changes in textile wastewater quality. In EC process, COD concentration of treated effluent was above the permitted direct discharge limits. So, active carbon was used to increase COD removal efficiency of EC process under the optimum operational conditions. COD and turbidity removal efficiencies in the EC/activated carbon (AC) process by adding 2,000 mg/l AC were 67.9 and 99.3%, respectively. Operation cost of EC/AC process is calculated as $1.64 \, \text{e/m}^3$. Besides, the operation costs at different operating conditions were also determined.

Keywords: Electrocoagulation; Taguchi method; Textile wastewater; Iron electrodes; COD removal; Turbidity removal

1. Introduction

Textile wastewaters with high chemical oxygen demand (COD) concentration, strong colour, high pH and temperature and low biodegradability are one of the significant pollutants for the environment [1,2]. Various physical, chemical and biological treatments have been widely used to treat the textile wastewater. Biological methods cannot be applied to most textile wastewaters due to the toxicity of most commercial

Presented at the 2nd International Conference on Recycling and Reuse (R&R2014), 4-6 June 2014, Istanbul, Turkey

1944-3994/1944-3986 © 2015 Balaban Desalination Publications. All rights reserved.

dyes to the organisms used in the process [3,4]. Chemical coagulation is not effective for the removal of dissolved reactive dyestuff [5]. Activated carbon (AC) adsorption has the associated cost and difficultly of the regeneration process and a high disposal cost. Advanced oxidation processes such as ozonation, UV and ozone/UV, photocatalysis and Fenton oxidation are not economically feasible [6,7]. Moreover, these conventional methods are also usually expensive and treatment efficiency is inadequate because of large variability in composition of textile wastewaters [2].

In recent years, electrochemical treatment methods such as electro-oxidation, electrocoagulation (EC) and electroflotation have attracted increasing attention for the treatment of various types of wastewaters. Electrochemical methods have been reported as a primary technique for treatment of various wastewaters by virtue of various benefits including environmental compatibility, versatility, energy efficiency, safety, selectivity, amenability to automation and cost effectiveness [2,8]. Electrochemical processes have some advantages such as simple equipment, easy operation, shortened retention time, rapid settling and decreased amount in precipitate or sludge.

Electrochemical methods were frequently used for treatment of wastewaters containing heavy metals [9,10], foodstuff [11], wastewaters of oil industries [12,13], textile industry [14,15], aqueous suspensions of ultrafine particles [16], nitrate [17], iron [18], phenolic compounds [19], arsenic and arsenate [20], refractory organic pollutants including lignin and EDTA [21], landfill leachate [22,23] and liquid organic fertilizer [24]. On the other hand, AC adsorption process has been used as an effective alternative technology for wastewater treatment. Pollutants are generally physically or chemically adsorbed onto the surface of the adsorbent. AC still remains as the most effective adsorbent. In several studies, combined adsorption and EC processes were used to enhance removal of pollutions and successful outcomes were obtained in textile wastewater treatment [25,26], removal of chromium from synthetic wastewater [27] and paper mill wastewater treatment [28].

EC treatment of textile dye solutions and/or wastewater was tested and fairly good removal efficiencies of COD, turbidity, colour and dissolved solids at varying operating conditions were obtained [2,29–31].

Recently, the statistical experimental designs have frequently been used in various fields of science, such as chemistry, engineering, agriculture and biology. In Taguchi method, experimental results are analysed to find the best or the optimal conditions for the product or the process. Besides keeping the experimental cost at a minimum level, another advantage of Taguchi method over the conventional experimental design methods is that it minimizes the variation in product response while keeping the mean response on target. Optimum working conditions determined from the laboratory work of Taguchi method can also be reproduced in the real production environment [32–35].

The present study was conducted to investigate the optimum operating conditions of textile wastewater treatment by EC process with iron electrodes and to determine the influence of the variables such as initial pH, current density and electrolysis time on treatment efficiency. The experiments were carried out in Taguchi orthogonal array (OA) experimental design (larger is better) with three levels and three factors. Besides, COD removal efficiencies with different amounts of AC were also investigated.

1.1. Removal mechanisms of EC

EC processes consist of metallic hydroxide flocs within the effluent to be cleaned through electrodissolution of soluble anodes. Aluminium and iron are the common electrode materials of EC. EC has three important processes: (1) electrolytic reaction at the surface of electrodes, (2) formation of coagulants in the aqueous phase and (3) adsorption of soluble or colloidal pollutants on coagulants and removal by sedimentation or floatation.

When the iron electrodes are used, the generated Fe^{3+} ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. For instance, ferric ions generated by electrochemical oxidation of iron electrode may form monomeric ions, $Fe(OH)_3$ and hydroxyl complexes namely: $Fe(H_2O)_6^{2+}$, $Fe(H_2O)_4^{2+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$ and $Fe_2(H_2O)_6$ $(OH)_4^{4+}$. Formation of these complexes depends strongly on solution pH [15,36]. Above pH > 9, Al(OH)^{4-} and Fe(OH)^{4-} are the dominant species.

When iron is used as electrode material, two mechanisms have been proposed for the production of $Fe(OH)_{n_V}$ where n = 2 or 3 [12,36].

$$Fe_{(s)} \rightarrow Fe^{2+}{}_{(aq)} + 2e^{-}(anode)$$
(1)

$$\operatorname{Fe}^{2+}_{(\mathrm{aq})} + 2\operatorname{OH}^{-}_{(\mathrm{aq})} \to \operatorname{Fe}(\operatorname{OH})_{2(\mathrm{s})}$$

$$\tag{2}$$

$$2H_2O_{(I)} + 2e^- \rightarrow H_{2(g)} + 2OH^-{}_{(aq)}(cathode) \eqno(3)$$

$$Fe_{(s)} + 2H_2O_{(I)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)}(\text{in solution})$$
(4)

$$\begin{array}{ll} 2Fe_{(s)} + 5H_2O_{(I)} + O_{2(g)} \rightarrow 2Fe(OH)_{3(s)} \\ &\quad + 2H_{2(g)}(\text{in solution}) \end{array} \tag{5}$$

2. Materials and methods

2.1. Wastewater source and characteristics

The wastewater samples were obtained from a textile factory in Cerkezkoy (Turkey). The factory has approximately 2,000 m³ of wastewater per day, has dyeing and finishing processes. Factory wastewater is directly discharged together with factory municipal wastewater.

The characteristics of wastewater used in the experiments are provided in Table 1.

2.2. EC reactor

The experimental set-up for the EC reactor was reported elsewhere [37]. The schematic diagram of electrochemical reactor is shown in Fig. 1. The EC reactor was made of Plexiglas with dimensions of $130 \times 100 \times 100$ mm. Four plate electrodes (two anodes and two cathodes) with dimensions of $50 \times 70 \times 2$ mm (purity $\geq 99.5\%$) were used in the study. The total effective electrode area was 210 cm² and the spacing between electrodes was 20 mm. The electrodes were connected to a digital DC power supply (Alpha 10A-50 V DC power supply) in monopolar-parallel mode.

2.3. Experimental procedure

The chemicals (AC, sodium hydroxide (NaOH) and hydrochloric acid (HCl)) were obtained from Merck. All of the experiments were carried out with 1,000 ml of wastewater at room temperature and 250 rpm mixing rate was used. Before each run, the wastewater was filtered through a filter paper having coarse pore sizes in order to get a homogenized particle size distribution in the sample. In the EC/AC

 Table 1

 Characteristics of wastewater used in the experiments

Parameter	Value	Discharge limit
pН	10.3	6–9
COD (mg/l)	780	250
BOD (mg/l)	375	-
Total solids (mg/l)	4,850	100
Conductivity (mS/cm)	10.34	-
Turbidity (NTU)	125	-

process, a desired amount of AC was added to the electrolytic reactor before electrical current density was turned on.

Electrodes were washed for 2 min by mixing with HCl and hexamethylenetetramine solutions after EC experiments. The electrodes were then rinsed through distilled water, dried, weighted and placed into the reactor. Then, again a filtration procedure was applied in order to eliminate a potential additional removal effect of filter paper, and chemical analyses were carried out. All investigated variables and their levels for EC are provided in Table 2.

The percentage removal efficiency of COD and turbidity was calculated using the following equation, Eq. (6).

Percentage removal efficiency(%) =
$$\left(\frac{(C_0 - C)}{C_0}\right) \times 100$$
(6)

where C_0 is the initial concentration and *C* is the final concentration of the pollutant (mg/l and NTU).

2.4. Analytical technique

COD, Biological oxygen demand (BOD) and Total solids (TS) were determined according to standard methods [38]. In all experiments, chemicals with analytic grade were used, 0.45 µm filter paper (Whatman, 47 mm) was used for TS measurements. COD of samples was determined using closed-reflux method. The pH, electrical conductivity and turbidity were measured using a pH metre (consort model C931), a WTW Model 340i conductivity metre and a HF model Micro TPI turbidimeter, respectively. The pH was adjusted by adding NaOH or HCl.

2.5. Operation cost

The operating cost is one of the most important parameters in the EC process because it effects the implementation of any method of wastewater treatment. The operating cost includes material (mainly electrodes) cost, electrical energy cost, as well as labour, maintenance and other costs. The latter costs items are largely independent on the electrode material [39–41]. Thus, in this study, the operating cost included only the costs of electrodes, electrical energy and chemicals. Calculation of operating cost is expressed as:

Operating Cost = A Energy_{consumption} + B Electrode_{consumption} + C Chemical_{consumption}

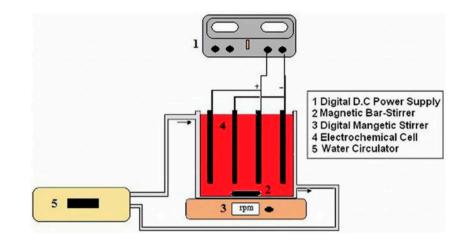


Fig. 1. Schematic diagram of experimental set-up.

Table 2 Operating factors and their levels

Factors	L1	L2	L3
A: Initial pH	7	8	9
B: Current density	75	100	150
C: Electrolysis time	10	20	30

where energy consumption and electrode consumption are consumption quantities per m³ of wastewater treated. Unit prices, A, B and C, given for the Turkish Market, September 2013, are as follows: electrical energy price $0.072 \in /kWh$, electrode material price $0.85 \in /kg$ for iron and chemical costs $0.73 \in /kg$ for NaOH, $0.29 \in /kg H_2SO_4$ and $60 \in /kg AC$.

The electrode ad energy consumptions in the EC process were calculated using the following equations:

$$Energy_{consumption} = \frac{(V \cdot I \cdot t)}{v}$$
(7)

where Energy_{consumption} is the energy consumption (kWh/m^3) , V is the voltage (Volts), I is the current (Amperes), t is the EC time (s) and v is the volume of the treated wastewater (m³). According to Faraday's laws, electrode material consumption and charge loading are calculated with the following equations;

$$\frac{\text{Faraday}}{\text{m}^3} = \frac{(I \cdot t)}{(F \cdot v)} \tag{8}$$

$$Electrode_{consumption} = \frac{(I \cdot t \cdot M_w)}{(z \cdot F \cdot v)}$$
(9)

where *F* is the Faraday's constant (96,485 C/mol), M_w is the molar mass of iron (56 g/mol) and *z* is the number of electron transfer (*z*Fe:2).

2.6 Statistical analysis

The variables chosen for this investigation are initial pH, current density and EC time. The Taguchi OA experimental design ($L_9(3^3)$ for three parameters each of with three values) was selected as the most suitable method for the experimental design. The experimental variables, their levels and results of conducted experiments are provided in Table 3.

In Taguchi robust design, the signal-to-noise (S/ N) ratio is used to monitor the performance measurements. The highest S/N ratios are the most preferred. Taguchi proposed more than 60 S/N ratios to be used in particular applications. The frequently used S/N ratio functions are "smaller is better", "larger is better" and "nominal is better". In general, a better signal is obtained when the noise is smaller, so a larger S/N ratio yields better final outcomes. "Larger is better" means that the desired output value for the optimized process is the maximum. In this situation, the S/N ratio can be calculated from (Eq. 10). According to the "smaller is better" situation, the desired value for the result of the process is the minimum. The S/N ratio for this situation can be calculated from Eq. (11). In the "nominal is best" situation, the desired response variable of optimized process is a nominal value. The performance characteristics were chosen as the optimization criteria. The "larger is better" was used to evaluate the system performance based on COD and turbidity removal efficiencies.

	Variables											
F	and their levels		uitial	Final	Removal	Initial	Final	f	- i	Electrode	Energy	Operation
Experiment no.	A B C	ງ <u>ອ</u> ບ	(mg/l)	(mg/l)	or CUD (%)	(NTU)	(NTU)	(%) turbidity	rinal pH	consump. (kg/m ³)	consump. (kWh/m ³)	(ϵ/m^3)
1	L1 L1 I	L1 78	780	505	35.3	125	78		7.9	0.27	5.90	0.67
2	L1 L2 I	L2 78	30	474	39.2	125	57.8		8.5	0.75	7.26	1.17
3	L1 L3 I	L3 78	30	451	40.2	125	2.3	98.1	9.6	1.71	6.90	1.97
4	L2 L1 I	L2 78	30	555	28.8	125	11.7		9.4	0.59	5.50	0.91
ъ С	L2 L2 I	L3 78	30	569	27.1	125	15.8		9.6	1.13	6.95	1.48
6	L2 L3 I	L1 78	30	596	23.6	125	7.8		9.5	0.57	8.43	1.11
7	L3 L1 I	L3 78	30	600	23.1	125	6.6		9.8	0.80	5.54	1.09
8	L3 L2 I	L1 78	30	646	17.2	125	24.5		9.6	0.45	5.77	0.81
6	L3 L3 I	L2 78	30	582	25.4	125	10.4		10.1	1.15	7.23	1.51

Experimental variables, their levels and results of conducted experiments corresponding to L₉ experimental design

Fable 3

Larger is better:

$$S/N = -10 \log_{10}\left(\frac{1}{n} \sum_{i:1}^{n} \frac{1^{2}}{Y_{i}}\right)$$
(10)

Smaller is better:

$$S/N = -10 \log_{10} \left(\frac{1}{n} \sum_{i:1}^{n} Y_{i}^{2} \right)$$
(11)

where *n* is the number of repetition done for an experimental combination and Y_i is the performance value of ith experiment. In Taguchi method, the experiment corresponding to optimum working conditions might not have been done during the whole period of the experimental stage. In such cases, the performance value corresponding to optimum working conditions can be predicted by utilizing the balanced characteristic of OA. For this, the additive model may be used [31,42].

$$Y_i = \mu + X_i + e_i \tag{12}$$

where μ is the overall mean of performance value, X_i is the fixed effect of the parameter level combination used in ith experiment and e_i is the random error in ith experiment. Because Eq. (12) is a point estimation, which is calculated using experimental data in order to determine whether results of the confirmation experiments are meaningful or not, the confidence interval must be evaluated. The confidence interval at a chosen error level may be calculated by [43].

$$Y_{i} \pm \sqrt{F_{\alpha;1,DF_{MS_{e}}}} MSe\left(\frac{1+m}{N} + \frac{1}{n_{i}}\right)$$
(13)

where *F* is the value of *F*-table, α is the error level, DF_{MSe} is the degrees of freedom of mean square error, *m* is the degrees of freedom used in the prediction of Y_i , *N* is the number of total experiments and n_i is the number of repetitions in the confirmation experiment. If experimental results are in percentage (%) before evaluating Eqs. (12) and (13), Ω transformation of percentage values should be applied first using the following equation. Values of interest are then later determined by carrying out reverse transformation using the same equation [31,44].

$$\Omega(db) = -10\log\left(\frac{1}{P} - 1\right) \tag{14}$$

where $\Omega(db)$ is the decibel value of the percentage value subjected to omega transformation and *P* is the percentage of the product obtained experimentally.

The order of the experiments was obtained by inserting parameters into columns of OA and L_3 (3³) was chosen as the experimental plan as given in Table 3. The order of experiments was made random in order to avoid noise sources which had not been considered initially and could occur during an experiment and affect results in a negative way.

3. Results and discussion

3.1. Optimum parametric levels

Experimental results of Taguchi model and intervals of variables are provided in Table 3. During the study, all of the experimental analysis carried out for both COD and turbidity results were performed in two repetitions.

The data collected from the experiments were analysed using the MINITAB (Trial Edition) software to assess the effect of each parameter on the optimization criterion. The obtained results are shown in Figs. 2 and 3. The numerical value of the maximum point in each graph shows the best value of that particular parameter (Table 3) and indicates the optimum conditions within the range of experimental conditions.

UV–vis scan results for EC process at different experimental conditions (experiments 1–9) are shown in Fig. 4. There was some pollution removal observed for EC process and the contaminations with strong adsorption peaks (between 200 and 400 nm) were removed by EC process. The lowest peak of the effluent was observed in experiment 3.

3.2. Effect of initial pH on EC process

The pH is an important parameter effecting removal efficiency of EC process [2,8]. To determine the effects of initial pH values on textile wastewater treatment efficiencies, the experiments provided in Table 3 were conducted. The effects of all performance criteria on COD removal efficiency were shown Fig. 2. The optimum pH value was seven (first level) for the best COD removal efficiency. The COD removal efficiencies slightly decreased as the initial pH increased from 7 to 9 (Fig. 2).

For turbidity removal, the best turbidity removal efficiency was obtained at a pH of 8 (second level). The initial pH values over 8 did not affect turbidity removal significantly.

3.3. Effect of current density on EC process

Current density is another important parameter on pollutant removal efficiency of EC process. Current density effects the metal hydroxide concentration formed and control the reaction rate during the process. Current density, especially, determines the coagulant dosage and bubble production, and hence affects the growth of flocs.

Effects of current density on textile wastewater treatment efficiency of EC process were studied in the range of $75-150 \text{ A/m}^2$. It is clear from Fig. 2 that increasing current densities did not increase COD removal efficiencies as it would be expected. Fig. 3 presents increasing turbidity removal efficiencies with increasing current densities. The best turbidity removal was obtained at level 3 (150 A/m^2).

3.4. Effect of electrolysis time on EC process

Electrolysis time is an important parameter with significant influences on EC process. Formation and amount of metal hydroxides play an important role in the removal of pollutants and such parameters are time-dependent parameters. Effect of electrolysis time on performance of EC process was investigated between 10 and 30 min.

As seen in Figs. 2 and 3, the removal efficiencies of COD increased with increasing operation time, but such an increase regressed after 20 min because of cathode reduction and formation of new electrocoagulant flocs. Effect of EC operation time was also investigated as another performance parameter. The highest turbidity removal efficiency was obtained at parametric level 3 (30 min).

3.5. Contribution ratios and statistical analysis

ANOVA was conducted to determine whether the process parameters were significant. The results are provided in Tables 4 and 5. As in the *S*/*N* ratio analysis, it is evident from Tables 4 and 5 that the significance of the factors prevails in the following order of importance: Initial pH > electrolysis time > current density for COD efficiencies and Initial pH > current density > electrolysis time for turbidity removal efficiencies. Firstly, in order to test the predicted results, confirmation experiments were carried out once at the same working conditions. Thus, some confirmation runs including the optimum working conditions were also made and presented in Table 6. Using Eq. (12), the Ω transformation was applied to the estimated results (predicted) to be able to express as a percentage of the

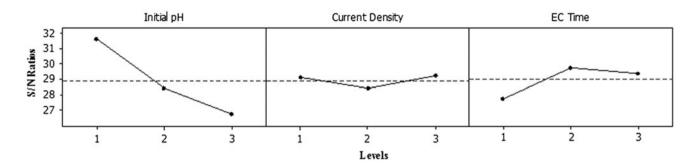


Fig. 2. The effect of each parameter on COD removal.

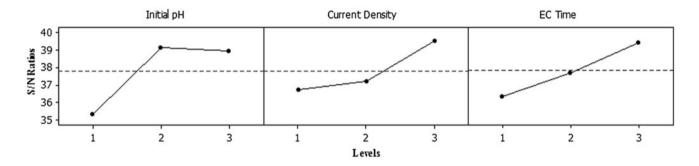


Fig. 3. The effect of each parameter on turbidity removal.

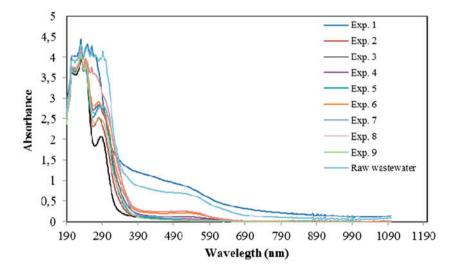


Fig. 4. Absorbance spectra of experiment effluents in EC process.

results given in Table 6. Moreover, contribution ratios (Cr) of each factor effecting COD and turbidity removal were determined (Tables 4 and 5).

According to Taguchi, the use of the F ratios in an ANOVA analysis is only helpful for the qualitative evaluation of whether factorial effects exist. For quan-

titative evaluation, this is something that can be achieved through the use of a Cr. The Cr of a main factor effect is its contribution (in percentage terms) to total variability of the experimental results [31,35]. The Cr can be achieved by dividing the source's net variation by SS_{total} (Eq. 15):

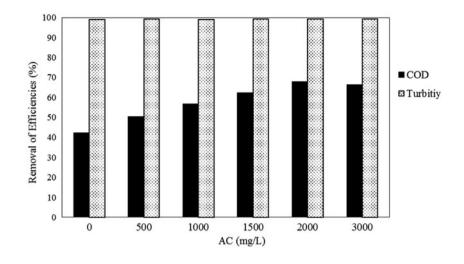


Fig. 5. Effect of AC on COD and turbidity removal efficiencies of the EC process.

$$Cr = \frac{SS - (DOF \times MS_{Error})}{SS_{total}}$$
(15)

The initial pH had the highest contributions to the variability of the experimental results. Contribution ratios on COD and turbidity were obtained as 87.06 and 44.3, respectively. For operation time, contribution ratios were 11.65 and 24.4, respectively. Current density had the lowest contribution ratios for COD but this contribution ratio for turbidity efficiencies was calculated as 25.5.

3.6. EC/AC process

The present results revealed that COD concentration of treated effluent was above the permitted direct-discharge limits (for COD 250 mg/l). So, AC was used to increase COD removal efficiency of EC process under the optimum operational conditions

Table 4	
Results of ANOVA for COD efficiencies	

Source	SS	DOF	MS	F	Cr
Initial pH	425.609	2	212.804	611.90	87.06
Current density	5.576	2	2.788	8.02	1.14
Electrolysis time	56.976	2	28.488	81.91	11.65
Error	0.696	2	0.348	-	0.15
Total	488.856	8	_	_	100

Note: SS: sum of squares; DOF: degree of freedom; Cr: percent contribution.

Table 5Results of ANOVA for turbidity efficiencies

SS	DOF	MS	F	Cr
1419.3	2	709.6	3.10	44.3
836.7	2	418.3	1.83	25.5
801.5	2	400.8	1.75	24.4
45.6	2	22.8	-	5.8
3103.1	8	-	-	100
	1419.3 836.7 801.5 45.6	1419.3 2 836.7 2 801.5 2 45.6 2	1419.3 2 709.6 836.7 2 418.3 801.5 2 400.8 45.6 2 22.8	1419.3 2 709.6 3.10 836.7 2 418.3 1.83 801.5 2 400.8 1.75 45.6 2 22.8 -

Note: SS: sum of squares; DOF: degree of freedom; Cr: percent contribution.

Table 6

Optimum experimental conditions predicted and observed removal efficiencies values

	optimal			ty l	
Parameter	Value	Level	Value	Level	
Initial pH (A)	7	1	2	8	
Current density (B)	150	3	3	150	
Electrolysis time (C)	20	2	3	30	
COD removal					
Observed (%)	42.241.3		98.1		
Predicted (%)			_a		
Turbidity removal					
Observed (%)	30.6		99.1		
Predicted (%)	_a		99.5		
Confidence (%)	39.2-43.	4	98.1-100		
Operating Cost (ϵ/m^3)	1.52		1.73		

^aunpredictable.

(for COD parameter: pH 7, 150 A/m^2 and 20 min operation time). Effects of AC on textile wastewater treatment efficiencies were studied in the range of 500-3,000 mg/L. As can be seen in Fig. 5, the highest COD removal efficiency (67.9% and 241 mg/l) was obtained from 2000 mg/L AC. Turbidity removal efficiency of the same treatment was 99.3%. Besides, the UV-Vis spectrum was used to detect the changes of textile wastewater quality. The results of absorbance spectra (190-1100 nm) of EC/AC and EC process are presented in Fig. 6. It was found that the contaminations with strong absorption peaks between 200-400 nm were removed by EC and EC/AC processes. There was some adsorption of contamination observed between 200 and 390 nm for EC/AC process (Fig. 6). Moreover, the colour spectrum bands (400-700 nm) of textile wastewater degraded uniformly during EC and

EC/AC processes. As seen in Fig. 7, the EC/AC process was fairly effective on the removal of colour. Similarly, Bellebia et al., investigated the removal of COD and Turbidity by combined EC and adsorption processes. In the EC process using iron and aluminium electrodes, COD removal efficiencies were obtained as 75.37% for Al and 78.76% for iron electrodes in 10 min operation time. In the same study, COD removal efficiencies by combined EC and adsorption processes were found to be 98.97% for aluminium at 120 min and 93.37% for iron at 180 min [28]. Narayanan and Ganesan investigated the removal of chromium from synthetic wastewater by EC and found EC and adsorption processes feasible. Researchers observed increased Cr removal with increasing adsorbent concentrations and obtained Cr levels compatible with discharge limits [27].

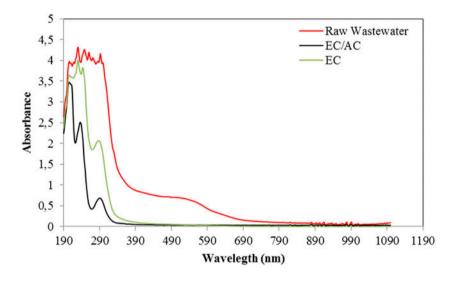
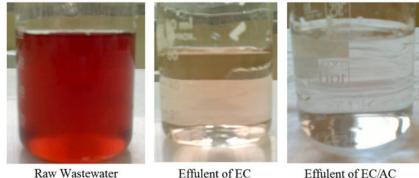


Fig. 6. Absorbance spectra of EC/AC and EC processes.



Raw Wastewater

Effulent of EC/AC

Fig. 7. Raw wastewater and effluent of EC and EC/AC processes.

4. Conclusions

In this study, an orthogonal Taguchi L_{9} (3³) array was employed to investigate the significance of EC parameters on COD and turbidity removal efficiencies. According to current results, the following conclusions were drawn: The initial pH was the most significant factor on COD and turbidity and its contribution ratio was 87.06 and 44.3%, respectively. The second most significant factor for COD is the electrolysis time and its contribution ratio was 11.63%. For turbidity, current density is the second signification factor and its contribution ratio was 25.5. Finally, the least effective process factor for COD was the current density and the least effective factor for turbidity was the electrolysis time. As a result of the statistical treatment, for COD removal, the defined set of optimal conditions was: Initial pH: 7, current density: 150 A/m^2 and electrolysis time: 20 min. On the other hand, for turbidity removal, optimal conditions were obtained as: initial pH: 8, current density: 150 A/m² and electrolysis time: 30 min. Operating cost of EC process for COD and turbidity were calculated as 1.52 and $1.73 \notin m^3$, respectively. In the EC/AC process, COD removal efficiency and operation cost of 2,000 mg/l AC treatment were obtained as 67.9% (241 mg/l) and calculated as 1.64 €/m³, respectively.

References

- S.H. Lin, M.L. Chen, Treatment of textile wastewater by chemical methods for reuse, Water Res. 31 (1997) 868–876.
- [2] M. Bayramoglu, M. Eyvaz, M. Kobya, Treatment of textile wastewater by electrocoagulation: Economical evaluation, Chem. Eng. J. 128 (2007) 155–161.
- [3] T. Robinson, G. McMullan, R. Marchant, P. Nigam, Remediation of dyes in textile effluent: A critical review on current treatment technologies with a proposed alternative, Bioresour. Technol. 77 (2001) 247–255.
- [4] O.T. Can, M. Kobya, E. Demirbas, M. Bayramoglu, Treatment of the textile wastewater by combined electrocoagulation, Chemosphere 62 (2006) 181–187.
- [5] P.C. Vandevivere, R. Bianchi, W. Verstraete, Treatment and reuse of wastewater from the textile wet-processing industry: Review of emerging technologies, J. Chem. Technol. Biotechnol. 72 (1998) 289–302.
- [6] S.F. Kang, C.H. Liao, M.C. Chen, Pre-oxidation and coagulation of textile wastewater by the Fenton process, Chemosphere 46 (2002) 923–928.
- [7] M. Koch, A. Yediler, D. Lienert, G. Insel, A. Kettrup, Ozonation of hydrolyzed azo dye reactive yellow 84 (CI), Chemosphere 46 (2002) 109–113.
- [8] M. Kobya, É. Demirbas, O.T. Can, M. Bayramoglu, Treatment of levafix orange textile dye solution by electrocoagulation, J. Hazard. Mater. 132 (2006) 183–188.

- [9] S. Vasudevan, J. Lakshmi, M. Packiyam, Electrocoagulation studies on removal of cadmium using magnesium electrode, J. Appl. Electrochem. 40 (2010) 2023–2032.
- [10] K. Ramakrishnan, G. Pandian, L. Jothinathan, V. Subramanyan, Removal of copper from water by electrocoagulation process-effect of alternating current (AC) and direct current (DC), Environ. Sci. Pollut. Res. 20 (2012) 399–412, doi: 10.1007/s11356-012-0855-7.
- [11] X. Chen, G. Chen, P.L. Yue, Separation of pollutants from restaurant wastewater by electrocoagulation, Sep. Purif. Technol. 19 (2000) 65–76.
- [12] U.B. Ogutveren, S. Koparal, Electrocoagulation for oilwater emulsion treatment, J. Environ. Sci. Health. 32A (1997) 2507–2520.
- [13] M.Y.A. Mollah, R. Schennach, J.P. Parga, D.L. Cocke, Electrocoagulation (EC)-science and applications, J. Hazard. Mater. 84 (2001) 29–41.
- [14] A.K. Golder, H. Kumar, A.N. Samata, S. Ray, Colour diminution and COD reduction in treatment of coloured effulent by electrocoagulation, Int. J. Environ. Eng. 2 (2010) 228–238.
- [15] A. Dalvand, M. Gholami, A. Joneidi, N.M. Mahmoodi, Dye removal, Energy consumption and operating cost of electrocoagulation of textile wastewater as a clean process, CLEAN–Soil Air Water 39 (2011) 665–672.
- [16] M.J. Matteson, R.L. Dobson, R.W. Glenn Jr, N.S Kukunoor, W.H. Waits III, E.J Clayfield, Electrocoagulation and separation of aqueous suspensions of ultrafine particles, Colloids Surf. A Physicochem. Eng. Aspects 104 (1995) 101–109.
- [17] S. Vasudevan, F. Epron, J. Lakshmi, S. Ravichandran, S. Mohan, G. Sozhan, Removal of NO₃⁻ from drinking water by electrocoagulation—An alternate approach, CLEAN-Soil Air Water 38 (2010) 225–229.
- [18] D. Ghosh, C.R. Medhi, M.K. Purkait, Treatment of drinking water containing iron using electrocoagulation, Int. J. Environ. Eng. 2 (2010) 212–227.
- [19] W. Phutdhawong, S. Chowwanapoonpohn, D. Buddhasukh, Electrocoagulation and subsequent recovery of phenolic compounds, Anal. Sci. 16 (2000) 1083–1084.
- [20] N. Balasubramanian, K. Madhavan, Arsenic removal from industrial effluent through electrocoagulation, Chem. Eng. Technol. 24 (2001) 519–521.
- [21] L.C. Chiang, J.E. Chang, S.C. Tseng, Electrochemical oxidation pretreatment of refractory organic pollutants, Water Sci. Technol. 36 (1997) 123–130.
- [22] M.O. Orkun, A. Kuleyin, Treatment Performance evaluation of chemical oxygen demand from landfill leachate by electro-coagulation and electro-fenton technique, Environ. Prog. Sustain. Energy 31 (2010) 59–67, doi: 10.1002/ep.10522.
- [23] M. Panizza, M. Delucchi, I. Sires, Electrochemical process for the treatment of landfill leachate, J. Appl. Electrochem. 40 (2010) 1721–1727.
- [24] A. Akyol, O.T. Can, E. Demirbas, M. Kobya, A comparative study of electrocoagulation and electrofenton for treatment of wastewater from liquid organic fertilizer plant, Sep. Purif. Technol. 112 (2013) 11–19.
- [25] S.-H. Chang, K.-S. Wang, H.-H. Liang, H.-Y. Chen, H.-C. Li, T.-H. Peng, Y.-C. Su, C.-Y. Chang, Treatment of reactive black 5 by combined electrocoagulation– granular activated carbon adsorption–microwave

regeneration process, J. Hazard. Mater. 175 (2010) 850–857.

- [26] M.S. Secula, B. Cagnon, T.F. Oliveria, O. Chedeville, H. Fauduet, Removal of acid dye from aqueous solutions by electrocoagulation/GAC adsorption coupling: Kinetics and electrical operation cots, J. Taiwan Inst. Chem. Eng. 43 (2012) 767–775.
- [27] N.V. Narayanan, M. Ganesan, Use of adsorption using granular activated carbon (GAC) for the enhancement of removal of chromium from synthetic wastewater by electrocoagulation, J. Hazard. Mater. 161 (2009) 575–580.
- [28] S. Bellebia, S. Kacha, A.Z. Bouyakoub, Z. Derriche, Experimental investigation of chemical oxygen demand and turbidity removal from cardboard paper mill effluents using combined electrocoagulation and adsorption processes, Environ. Prog. Sustain. Energy 31 (2012) 361–370.
- [29] N. Daneshvar, H.A. Sorkhabi, A. Tizpar, Decolorization of orange II by electrocoagulation method, Sep. Purif. Technol. 21 (2003) 153–162.
- [30] A. Gurses, M. Yalcın, C. Dogar, Electrocoagulation of some reactive dyes: A statistical investigation of some electrochemical variables, Waste Manage. 22 (2002) 491–499.
- [31] O. Gokkus, Y.S. Yıldız, B. Yavuz, Optimization of chemical coagulation of real textile wastewater using Taguchi experimental design method, Desalin. Water Treat. 49 (2012) 263–271.
- [32] L.I. Tong, C.H. Wang, C.C. Chen, C.T. Chen, Dynamic multiple responses by ideal solution analysis, Eur. J. Oper. Res. 156 (2004) 433–444.
- [33] J.J. Pignatiello, An overview of the strategy and tactics of taguchi, IIE Transactions 20 (1988) 247–254.
- [34] G. Taguchi, S. Choudhury, Y. Wu, Quality Engineering: The Taguchi method, in: Taguchi's Quality Engineering Handbook, John Wiley & Sons, New Jersey, NJ, 2004, ISBN 9780-471-4133-49.

- [35] S.Y. Yıldız, Optimization of Bomaplex Red CR-L dye removal from aqueous solution by electrocoagulation using aluminium electrodes, J. Hazard. Mater. 153 (2008) 194–200.
- [36] N. Daneshvar, H. Ashassi-Sorkhabi, M.B. Kasiri, Decolorization of dye solution containing Acid Red 14 by electrocoagulation with a comparative investigation of different electrode connections, J. Hazard. Mater. B 112 (2004) 55–62.
- [37] F. Ozyonar, B. Karagozoglu, Investigation of technical and economic analysis of electrocoagulation process for the treatment of great and small cattle slaughterhouse wastewater, Desalin. Water Treat. 52 (2014) 74–87.
- [38] APHA, AWWA, EF, Standard Methods for the Examination of Water and Wastewater, twentieth ed., American Public Health Association, Washington, DC, 1998.
- [39] J.C. Donini, J. Kan, J. Szynkarczuk, T.A. Hassan, K.L. Kar, The operating cost of electrocoagulation, Canadian J. Chem. Eng. 72 (1994) 1007–1012.
- [40] M. Kobya, H. Hiz, E. Senturk, C. Aydiner, E. Demirbas, Treatment of potato chips manufacturing wastewater by electrocoagulation, Desalination 190 (2006) 201–211.
- [41] M. Bayramoglu, M. Kobya, M. Eyvaz, E. Senturk, Technical and economic analysis of electrocoagulation for the treatment of poultry slaughterhouse wastewater, Sep. Purif. Technol. 51 (2006) 404–408.
- [42] M.S. Phadke, R.N. Kackar, D.D. Speeney, M.J. Grieco, Quality control, robust design, and the Taguchi method: off-line quality control in integrated circuit fabrication using experimental design, Springer, US, 1998, pp. 99–141.
- [43] P.J. Ross, Taguchi Techniques for Quality Engineering. McGraw-Hill, New York, NY, 1988, pp. 123–125.
- [44] G. Taguchi, System of Experimental Design, Quality Resources, American Supplier Institute, Dearborn, MI, 1987, pp. 108–115.