

# The use of artificial neural networks (ANN) for modeling of decolorization of textile dye solution containing C. I. Basic Yellow 28 by electrocoagulation process

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## Abstract

In this paper, electrocoagulation has been used for removal of color from solution containing C. I. Basic Yellow 28. The effect of operational parameters such as current density, initial pH of the solution, time of electrolysis, initial dye concentration, distance between the electrodes, retention time and solution conductivity were studied in an attempt to reach higher removal efficiency. Our results showed that the increase of current density up to  $80 \text{ A m}^{-2}$  enhanced the color removal efficiency, the electrolysis time was 7 min and the range of pH was determined 5–8. It was found that for achieving a high color removal percent, the conductivity of the solution and the initial concentration of dye should be  $10 \text{ mS cm}^{-1}$  and  $50 \text{ mg l}^{-1}$ , respectively. An artificial neural networks (ANN) model was developed to predict the performance of decolorization efficiency by EC process based on experimental data obtained in a laboratory batch reactor. A comparison between the predicted results of the designed ANN model and experimental data was also conducted. The model can describe the color removal percent under different conditions.

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**Keywords:** Artificial neural networks; Electrocoagulation; Modeling; Decolorization; C. I. Basic Yellow 28

## 1. Introduction

Many industries such as plastics, paper, textile and cosmetics use dyes in order to color their products. These molecules are common water pollutants and they may be frequently found in trace quantities in industrial wastewaters. Textile plants, particularly those involved in finishing processes, are major water consumers and the source of considerable pollution. The disposal of these colored wastewaters poses a major problem for the industry as well as a threat to the environment. There are many processes to remove dyes from colored effluents such as adsorption, precipitation, chemical degradation, photodegradation, biodegradation, chemical coagulation and electrocoagulation [1–3]. Adsorption and precipitation processes are very time-consuming and costly with low efficiency. Chemical degra-

ation by oxidative agents such as chlorine is the most important and effective methods, but it produces some very toxic products such as organochlorine compounds [1]. Photooxidation by UV/H<sub>2</sub>O<sub>2</sub> or UV/TiO<sub>2</sub> needs additional chemicals and therefore causes a secondary pollution. Although biodegradation process is cheaper than other methods, it is less effective because of the toxicity of dyes that has an inhibiting effect on the bacterial development [2,3].

Hence, electrocoagulation (EC) as an electrochemical method was developed to overcome the drawbacks of conventional water and wastewater treatment technologies. EC process provides a simple, reliable and cost-effective method for the treatment of wastewater without any need for additional chemicals, and thus the secondary pollution. It also reduces the amount of sludge, which needs to be disposed [3–5].

EC technique uses a direct current source between metal electrodes immersed in polluted water. The electrical current causes the dissolution of metal electrodes commonly iron or aluminum into wastewater. The metal ions, at an appropriate pH, can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate the suspended particles or precipitate

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### Nomenclature

ANN	artificial neural networks
BY28	C. I. Basic Yellow 28
$C_0$	initial concentration of dye ( $\text{mg l}^{-1}$ )
CD	current density ( $\text{A m}^{-2}$ )
CR (%)	color removal percent (%)
DE	distance between the electrodes (mm)
RTS	retention time of sludge (min)
$t_{\text{EC}}$	time of electrolysis (min)

### Greek letter

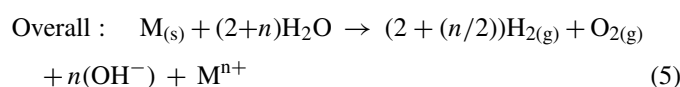
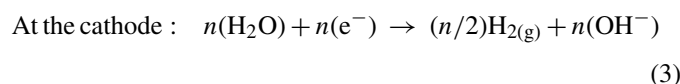
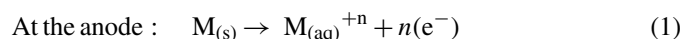
$\gamma$	conductivity ( $\text{mS cm}^{-1}$ )
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and adsorb dissolved contaminants [6]. The most widely used electrode materials in EC process are aluminum and iron [7].

Generally, six main processes occur during electrocoagulation:

1. Migration to an oppositely charged electrode (electrophoresis) and aggregation due to charge neutralization.
2. The cation or hydroxyl ion ( $\text{OH}^-$ ) forms a precipitate with the pollutant.
3. The metallic cation interacts with  $\text{OH}^-$  to form a hydroxide, which has high adsorption properties thus bonding to the pollutant (bridge coagulation).
4. The hydroxides form larger lattice-like structures and sweeps through the water (sweep coagulation).
5. Oxidation of pollutants to less toxic species.
6. Removal by electroflotation or sedimentation and adhesion to bubbles [3–6].

Interactions occurring within a batch electrocoagulation reactor are shown in Fig. 1 [6]. Main electrochemical reactions at the electrodes are [8]:



when  $\text{M}_{(\text{s})} = \text{Fe}$  metal is used as anodes, iron upon oxidation in an electrolytic system produces  $\text{Fe}(\text{OH})_2^{2+}$ ,  $[\text{Fe}(\text{OH})_2]^{+}$ ,  $[\text{Fe}(\text{OH})_2]^{4+}$ ,  $[\text{Fe}(\text{OH})_4]^{-}$ ,  $[\text{Fe}(\text{H}_2\text{O})_2]^{+}$ ,  $[\text{Fe}(\text{H}_2\text{O})_5\text{OH}]^{2+}$ ,  $[\text{Fe}(\text{H}_2\text{O})_4(\text{OH})_2]^{+}$ ,  $[\text{Fe}(\text{H}_2\text{O})_8(\text{OH})_2]^{4+}$ ,  $[\text{Fe}_2(\text{H}_2\text{O})_6(\text{OH})_4]^{2+}$ , which transform finally into  $\text{Fe}(\text{OH})_3$  or  $\text{Fe}(\text{OH})_2$  form [4,7].

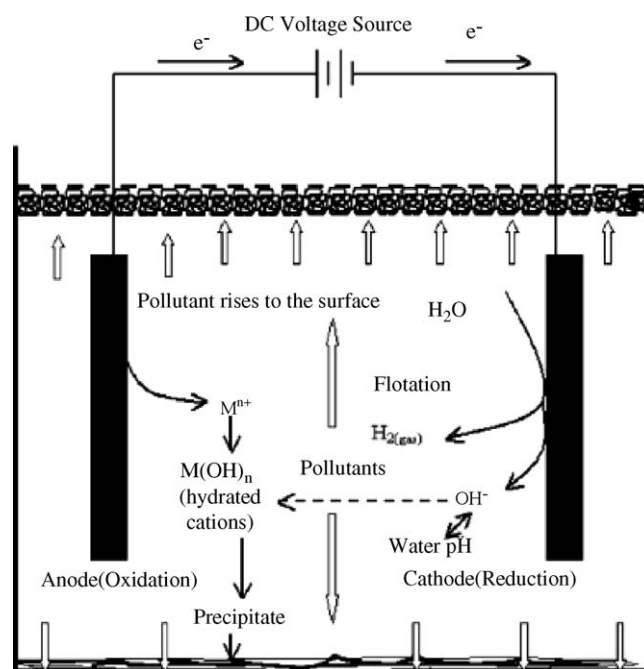


Fig. 1. Schematic diagram of a bench-scale two-electrode electrocoagulation cell.

Due to the complexity of the reactions in the electrocoagulation systems, the kinetic parameters of the various steps involved are very difficult to determine, leading to uncertainties in the design and scale-up of chemical reactors of industrial interest. To the best of our knowledge, kinetic modeling of electrocoagulation processes has been little investigated. Recently Carmona et al. [9] developed a model for prediction of treatment of oil suspensions. The adsorption equilibrium of organic matter on Al hydroxide was modeled using three kinetic equations. Lapique et al. [10] proposed a phenomenological model for treatment of industrial wastes by electrocoagulation, taking into account the latency period and based on global complexation equilibrium between Al(III) species and the matter.

Artificial neural networks (ANN) are a promising alternative modeling technique. One of the characteristics of modeling based on artificial neural networks is that it does not require the mathematical description of the phenomena involved in the process, and might therefore prove useful in simulating and up-scaling complex electrocoagulation systems. The success in obtaining a reliable and robust network depends strongly on the choice of process variables involved as well as the available set of data and the domain used for training purposes [11].

In this study, decolorization of the Textile Methine dye solution containing BY28 by electrocoagulation method has been investigated. This dye is soluble in water and belongs to cationic basic dyes group. It is idoneous for acrylic fiber dyeing and used in the wool and blanket factories. The performance of electrocoagulation method for removing of color from this dye solution has been evaluated. The effects of operational parameters such as current density, initial pH, electrolysis time, distance between the electrodes, retention time, dye concentration and solution conductivity on color removal efficiency have been also investigated. An important objective was to obtain an ANN model that

could make reliable prediction of the efficiency of the electrocoagulation process.

## 2. Experimental

### 2.1. Materials and methods

The commercial dye used in this project, was purchased from Bezema (Switzerland). The chemical structure and other characteristics of this dye are shown in Table 1. Dye solution was prepared by dissolving the dye in distilled water. The conductivity of solution was raised up and adjusted to different values by addition of NaCl (Fluka, Switzerland). The conductivity measurement was performed using a Philips conductimeter (PW 9509, England). The pH of the solution was measured by pH meter (Metrohm 654, Switzerland) and adjusted by adding NaOH (1 N) or H<sub>2</sub>SO<sub>4</sub> (1 N) (Merck, Germany) solutions. Iron (ST 37-2) plates were used as anode and cathode. Dimensions of electrodes were 50 mm × 50 mm × 3 mm in all experiments. The electrodes were connected to a DC power supply (ADAK PS808, Iran) with galvanostatic operational option. All the runs were performed at room temperature. In each run, 250 ml of the dye solution was decanted into the electrolytic cell. The current density was adjusted to a desired value and the electrolysis was started. At the end of electrocoagulation, all samples were allowed to settle in a 250 ml vessel before any analysis.

### 2.2. Chemical analysis

The dye concentration was determined from its absorbance characteristic in the UV–vis range (200–800 nm) with the calibration method. A WPA lightwave (S2000, England) spectrophotometer connected to a PC was used. For this measurement the maximum absorption ( $\lambda_{\max}$ ) wavelength of the dye was determined by measurement of their absorbance at various wavelengths (Table 1). The calculation of color removal efficiency after electrocoagulation treatment was performed.

The chemical oxygen demand (COD) of dye solutions was measured according to the standard methods for examining water and wastewater (Ampoule method) [12].

### 2.3. ANN software

All ANN calculations carried out using Matlab 6.5 mathematical software with ANN toolbox for windows running on a

personal computer (Pentium IV 2800 MHz). A three-layer network with a sigmoidal transfer function (*trainscg*) with back propagation algorithm was designed in this study.

## 3. Results and discussion

### 3.1. Neural network modeling

ANNs are direct inspiration from the biology of human brain, where billions of neurons are interconnected to process a variety of complex information. Accordingly, a computational neural network consists of simple processing units called neurons [13–15]. In general, a neural net (multilayered perceptron), as shown in Fig. 2, is parallel interconnected structure consisting of: (1) input layer of neuron (independent variables), (2) a number of hidden layers, (3) and output layer (dependent variables). The number of input and output neurons is fixed by the nature of the problem. The hidden layers act like feature detectors there can be more than one hidden layer. Universal approximation theory, however, suggests that a network with a single hidden layer with a sufficiently large number of neurons can interpret any input–output structure [11].

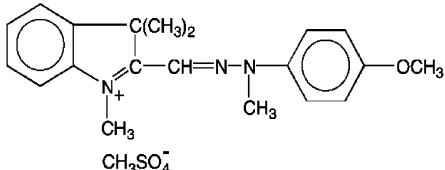
The topology of an artificial neural network (ANN) is determined by the number of layers in the ANN, the number of nodes in each layer and the nature of the transfer functions. Correct identification of the set of independent input variables and the output variables is the first task in building ANN model for a process. Optimization of ANN topology is probably the next important step in the development of a model. We used three-layered feed forward back propagation neural network (7:10:1) for modeling of EC Process (Fig. 2). In the present work, the input variables to the feed forward neural network were as follows: the current density (A m<sup>-2</sup>), electrolysis time (min), initial pH, initial dye concentration (mg l<sup>-1</sup>), conductivity (mS cm<sup>-1</sup>), retention time of sludge (min) and distance between electrodes (mm). Color removal percent (CR (%)) was chosen as the experimental response or output variable.

In this work, the sigmoidal transfer function was used as a transfer function in the hidden and output layers. This is the most widely used transfer function, which is given by:

$$f(x) = \frac{1}{1 + \exp(-x)} \quad (6)$$

where  $f(x)$  is the hidden neuron output [11,13]. The training function was *trainscg*. Out of the several data points generated, 49 experimental sets were used to develop the ANN model. The

Table 1  
Characteristics of the commercial dye

Structural	Commercial name	C. I. number	$\lambda_{\max}$ (nm)	Chemical class	$M_w$ (g mol <sup>-1</sup> )
	Golden Yellow X-GL 200%	48054	438	Methine	433

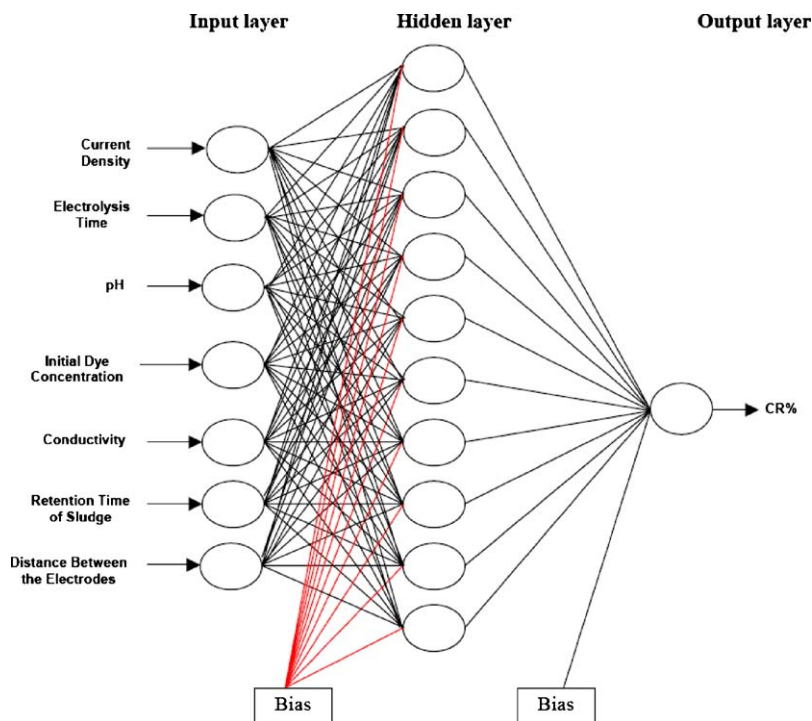


Fig. 2. The ANN optimized structure.

range of variables studied is summarized in Table 2. The samples were split into training, validation and test subsets that each of them contains 25, 12 and 12 samples, respectively. The validation and test sets, for evaluation of the validation and modeling power of the model, were randomly selected from the experimental data. Since the used transfer function in the hidden layer was sigmoid, all samples were scaled in the 0.2–0.8 ranges. So any samples ( $X_i$ ) (from the training, validation and test sets) were scaled to a new value  $A_i$  as follows:

$$A_i = 0.2 + \frac{0.6[X_i - \min(X_i)]}{\max(X_i) - \min(X_i)} \quad (7)$$

where  $\min(X_i)$  and  $\max(X_i)$  are the extreme values of variable  $X_i$  [16].

In order to calculate training, validation and test errors all of the outputs were performed an inverse range scaling to return the predicted responses to their original scale and compared them with experimental responses. Fig. 3 shows a comparison

between calculated and experimental values of the output variable (CR (%)), using the neural network model with number of hidden layer equal to 10. We used two lines to show the success of the prediction. The one is the perfect fit (predicted data equal to experimental data), on which all the data of an ideal model should lay. The other line is the line that best fits on the data of the scatter plot with equation  $Y = ax + b$  and it is obtained with regression analysis based on the minimization of the squared errors. The correlation coefficient of this line is also presented ( $R^2$ ). The closer to 1 this factor is and the closer the coefficients of the line to 1 and 0, respectively, are the better the model is. The plot in this figure has correlation coefficient of 0.974 for the test

Table 2  
Model variables and their ranges

Variable	Range
<b>Input layer</b>	
Current density	20–120 A m <sup>-2</sup>
Electrolysis time	2–7 min
Initial pH	2.1–11.2
Initial dye concentration	40–200 mg l <sup>-1</sup>
Conductivity	3.1–21.1 mS cm <sup>-1</sup>
Retention time of sludge	5–40 min
Distance between electrodes	5–30 mm
<b>Output layer</b>	
Color removal percent	0–100%

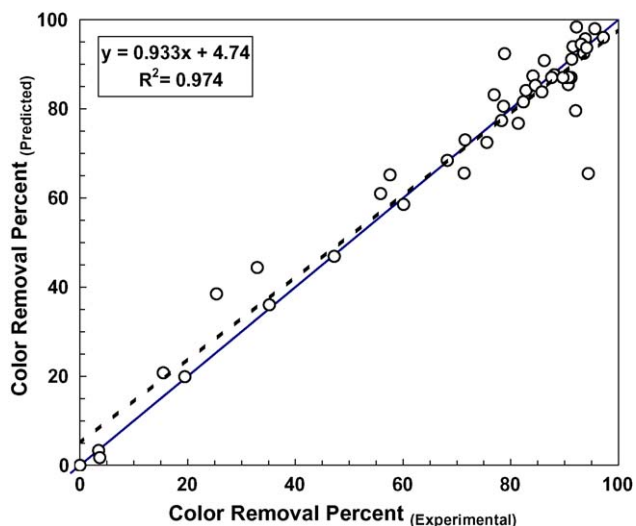


Fig. 3. Comparison of the experimental results with those calculated via neural network modeling.

set. These results confirm that the neural network model reproduces the decolorization in our system, within the experimental ranges adopted in the model fitting.

### 3.2. Effect of current density on the efficiency of color removal

In all electrochemical processes, current density determines the coagulant production rate, and adjusts the rate and size of the bubble production and hence affects the growth of flocs [17,18]. To investigate the effect of current density on the efficiency of color removal, electrocoagulation process was carried out using various current densities.

With Fig. 4 shows a comparison between calculated and experimental values of the output variable (CR (%)) as a function of current density using the neural network model with 10 neurons in the hidden layer. The agreement between the model predictions and the experimental data is good. Increasing of current density causes the increase of oxidized iron production from electrodes. The optimum current density of  $80 \text{ A m}^{-2}$  was used for the color removal from dye solution.

### 3.3. Effect of time of electrolysis on the efficiency of color removal

Reaction time influences the treatment efficiency of the electrochemical process. Electrolysis time ( $t$ ) determines the rate of production of  $\text{Fe}^{2+}$  or  $\text{Fe}^{3+}$  ions from iron electrodes [3]. Fig. 5 shows the relationship between the color removal efficiency and the electrolysis time. The color removal efficiency depends directly on the concentration of hydroxyl and metal ions produced on the electrodes. According to the results showed in Fig. 5, the optimum electrolysis time was 7 min for the color removal from dye solution. Fig. 5 there and simply mentions that it shows fair agreement between predictions from ANN model and experimental results. From this plot it can be seen that obtained results from the proposed

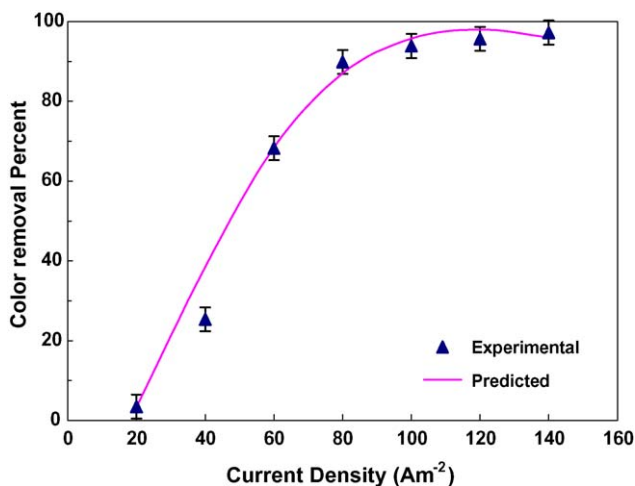


Fig. 4. Comparison between ANN predicted and experimental values of CR (%) as a function of current density ( $C_0 = 50 \text{ mg l}^{-1}$ ,  $t_{\text{EC}} = 7 \text{ min}$ ,  $\text{RTS} = 30 \text{ min}$ ,  $\text{DE} = 15 \text{ mm}$ ,  $\text{pH} = 5.8$  and  $\gamma = 10 \text{ mS cm}^{-1}$ ).

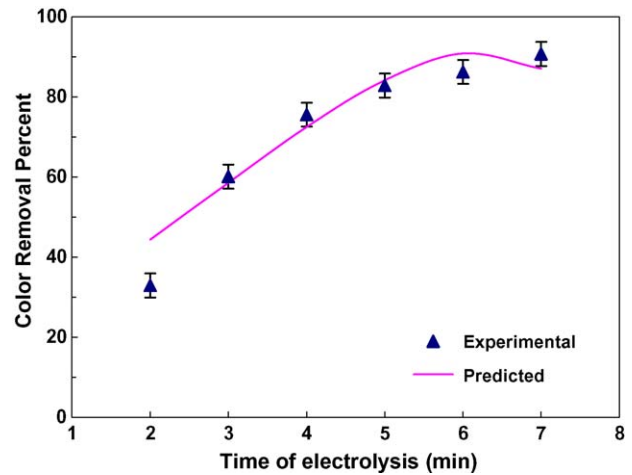


Fig. 5. Comparison between ANN predicted and experimental values of CR (%) as a function of electrolysis time ( $\text{CD} = 80 \text{ A m}^{-2}$ ,  $C_0 = 50 \text{ mg l}^{-1}$ ,  $\text{RTS} = 30 \text{ min}$ ,  $\text{DE} = 15 \text{ mm}$ ,  $\text{pH} = 5.8$  and  $\gamma = 10 \text{ mS cm}^{-1}$ ).

ANN model are in good agreement with the experimental data.

### 3.4. Effect of initial pH on the efficiency of color removal

It has been established that the influent pH is an important parameter influencing the performance of the EC process [18]. To examine its effect, the dye solution was adjusted to the desired pH for each experiment by adding sodium hydroxide or sulfuric acid solution. The experimental and ANN calculated values of color removal percent for dye solution with various initial pH values were shown in Fig. 6. The experimental results showed that when pH of the dye solution was between 5 and 8, there was maximum color removal efficiency. As observed by other investigators [19,20], a pH increase occurs when the solution pH is low. However, when the solution pH is above 9, a pH decrease occurs. In other words, electrocoagulation can act as a pH neutralizer [19]. As the initial pH values of this dye solution were around 5.8, there was no need for addition of chemicals to change the initial pH value.

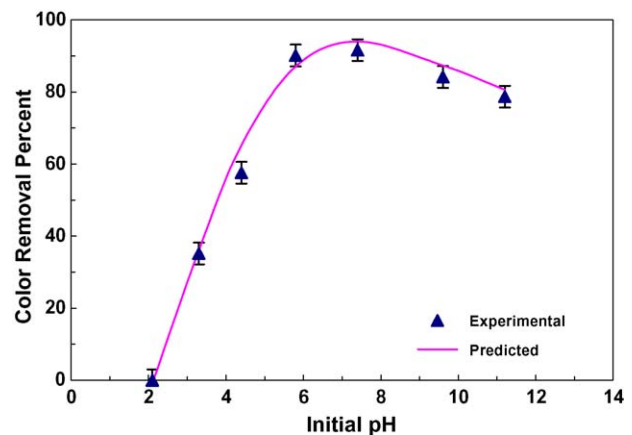


Fig. 6. Comparison between ANN predicted and experimental values of CR (%) as a function initial pH ( $C_0 = 50 \text{ mg l}^{-1}$ ,  $\text{CD} = 80 \text{ A m}^{-2}$ ,  $t_{\text{EC}} = 7 \text{ min}$ ,  $\text{RTS} = 30 \text{ min}$ ,  $\text{DE} = 15 \text{ mm}$  and  $\gamma = 10 \text{ mS cm}^{-1}$ ).

### 3.5. Effect of initial dye concentration on the efficiency of color removal

The dye solution with different initial concentrations in the range of 20–200 mg l<sup>-1</sup> was treated by EC in optimized current density and time of electrolysis values. The experimental and ANN predicted values of color removal percent was plotted against related initial dye concentration (Fig. 7). Up to the concentration of 50 mg l<sup>-1</sup>, the adsorption capacity of flocs was not exhausted and the rate of color removal was relatively constant. However, beyond this concentration, the adsorption capacity of flocs became exhausted, especially when it was more than 80 mg l<sup>-1</sup>.

### 3.6. Effect of the conductivity on the efficiency of color removal

The sodium chloride concentration was varied to evaluate the impact of solution conductivity on electrocoagulation in this section. It should be noted that the conductivity of the solution affects the current efficiency, cell voltage and consumption of electrical energy in electrolytic cells [3]. In order to examine the effect of conductivity on decolorization, NaCl was added to solution to adjust its conductivity. It appeared that conductivity had little effect on treatment efficiencies in the investigated range from 2 to 21 mS cm<sup>-1</sup> (Fig. 8). According to the results, high color removal percentage with low cell voltages and low energy consumption can be obtained in dye solutions with a conductivity of around 10 mS cm<sup>-1</sup>. Fig. 8 shows the predicted CR (%) estimated from the ANN model (solid line) and experimental ones as a function of solution conductivity. It can be seen that the ANN model correctly predicts the trend of the CR (%).

### 3.7. Effect of distance between the electrodes on the removal efficiency

The variation of experimental and ANN calculated values of color removal efficiency during the reaction period under dif-

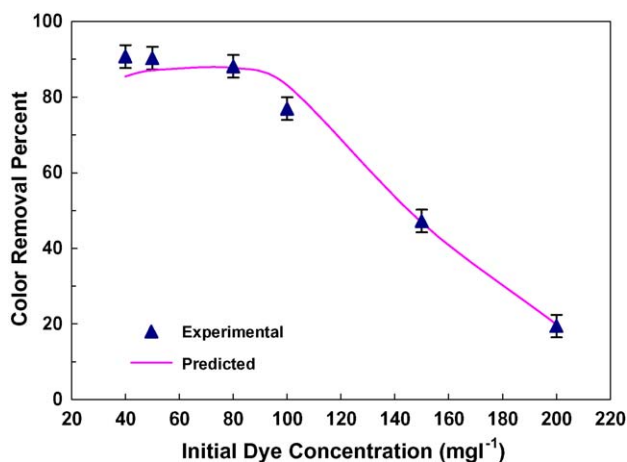


Fig. 7. Comparison between ANN predicted and experimental values of CR (%) as a function of electrolysis time initial concentration of dye (CD = 80 A m<sup>-2</sup>,  $t_{EC}$  = 7 min, RTS = 30 min, DE = 15 mm, pH 5.8 and  $\gamma$  = 10 mS cm<sup>-1</sup>).

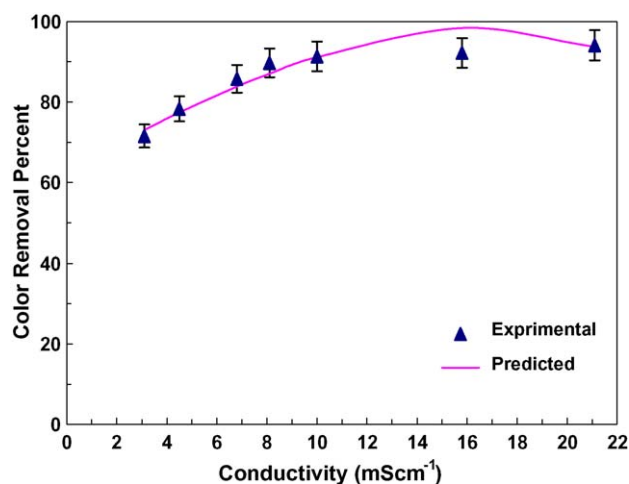


Fig. 8. Comparison between ANN predicted and experimental values of CR (%) as a function of conductivity of solution ( $C_0$  = 50 mg l<sup>-1</sup>, CD = 80 A m<sup>-2</sup>,  $t_{EC}$  = 7 min, RTS = 30 min, pH 5.8 and DE = 15 mm).

ferent iron plants (electrode distance) are presented in Fig. 9. According to the results for a solution with a dye concentration of 50 mg l<sup>-1</sup>, the maximum efficiency of color removal was observed at interelectrode distance 15 mm. So, the optimal electrode distance is 15 mm for this equipment in consideration of the efficiency together.

### 3.8. Effect of retention time on the removal efficiency

Once the electrolysis process was performed, the samples were poured into graduated cylinders for precipitation of floc. When retention time is increased (up to 25 min), the removal efficiency of BY28 is slightly increased. After 30 min, adsorbed dyes from dye solution, was almost constant. The experimental and ANN predicted values of color removal efficiency were shown in Fig. 10.

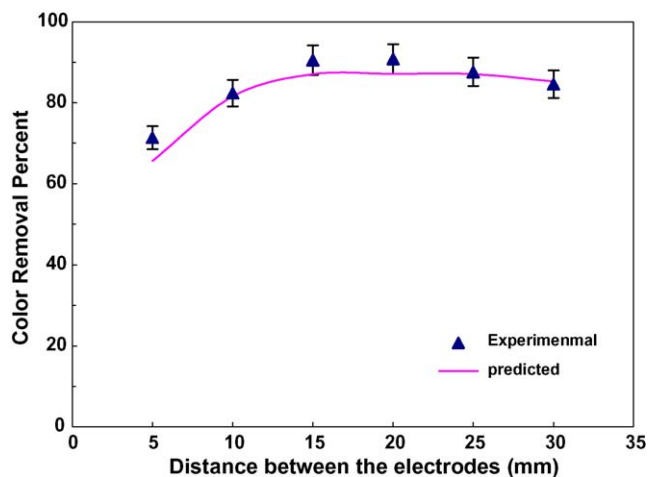


Fig. 9. Comparison between ANN predicted and experimental values of CR (%) as a function of distance between the electrodes ( $C_0$  = 50 mg l<sup>-1</sup>, CD = 80 A m<sup>-2</sup>,  $t_{EC}$  = 7 min, RTS = 30 min, pH 5.8 and  $\gamma$  = 10 mS cm<sup>-1</sup>).

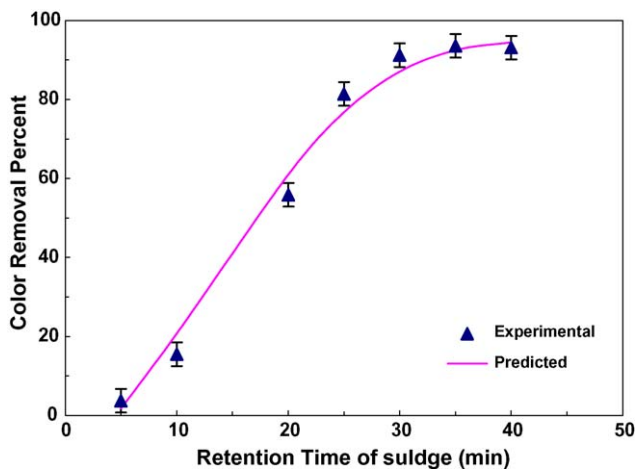


Fig. 10. Comparison between ANN predicted and experimental values of CR (%) as a function of retention time of sludge ( $C_0 = 50 \text{ mg l}^{-1}$ ,  $CD = 80 \text{ A m}^{-2}$ ,  $t_{EC} = 7 \text{ min}$ ,  $DE = 15 \text{ mm}$ ,  $\text{pH} = 5.8$  and  $\gamma = 10 \text{ mS cm}^{-1}$ ).

### 3.9. Absorbance spectra and COD reduction of the dye solution

Fig. 11 illustrates the absorption spectra of dye solution before and after EC process using optimized experimental value. It can be seen that EC process in the optimized condition causes near complete removal of color from dye solution. Then, subsequent to decolorization in optimized condition ( $C_0 = 50 \text{ mg l}^{-1}$ , current density  $80 \text{ A m}^{-2}$  and electrolysis time of 7 min), the COD of the treated solutions was measured again. The COD was reduced more than 65%.

### 3.10. Operation cost in optimum condition

Operation cost is very important economical parameters in EC process like all other electrolytic processes. The operating cost includes material cost (mainly electrodes), utility cost (mainly electrical energy), as well as labor, maintenance and other fixed costs. In this preliminary economic study, energy and electrode material costs are taken into account as major cost

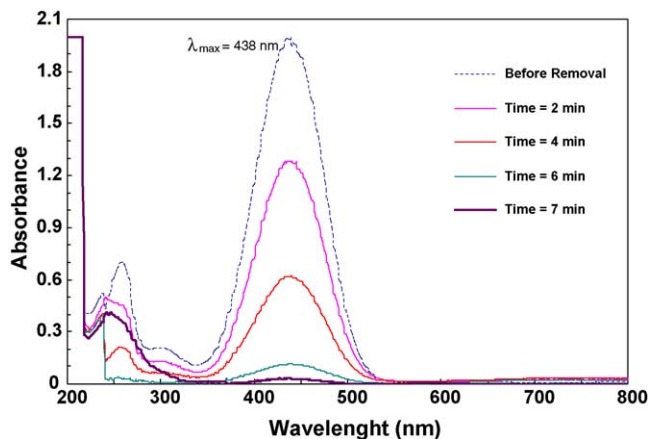


Fig. 11. Absorbance spectra of BY28, recorded before EC and after at different electrolysis times in the optimized condition:  $C_0 = 50 \text{ mg l}^{-1}$ ,  $CD = 80 \text{ A m}^{-2}$ ,  $\text{RTS} = 30 \text{ min}$ ,  $DE = 15 \text{ mm}$ ,  $\text{pH} = 5.8$  and  $\gamma = 10 \text{ mS cm}^{-1}$ .

items in the calculation of the operating cost as kWh/kg of dye removed [3]:

$$\text{Operating cost} = aC_{\text{energy}} + bC_{\text{electrode}} \quad (8)$$

where  $C_{\text{energy}}$  and  $C_{\text{electrode}}$ , are consumption quantities per kg of dye removed, which are obtained experimentally. Given Iranian market in May 2006, unit prices  $a$  and  $b$  are as follows:

- (a) electrical energy price  $0.0068 \text{ US\$/kWh}$ ;
- (b) electrode material price  $0.3 \text{ US\$/kg}$ .

These calculations were carried out after optimizing the operational parameters in EC process. The calculated value was obtained around  $0.07 \text{ US\$}$  for each kg dye removed.

## 4. Conclusion

Electrocoagulation process was used for removal of color from dye solution containing C. I. Basic Yellow 28. The effect of various operational parameters on color removal efficiency was investigated and optimized. Results showed that applying optimum current density of  $80 \text{ A m}^{-2}$  could remove a high percent of color from dye solution. It was found that the proper electrolysis time for the removal of color from this dye solution was 7 min. The dye solution decolorized more efficiency when the initial pH value of the solution was between 5 and 8. Color removal efficiency by EC decreased when the initial dye concentration was more than  $80 \text{ mg l}^{-1}$ . The complete removal of color, after selection of optimal operational parameters, could be achieved in a relatively short time of about 7 min. Artificial neural network modeling has been successfully used to investigate the cause effect relationship in electrocoagulation process. The ANN model could describe the behavior of the complex reaction system with the range of experimental conditions adopted. Simulation based on the ANN model can estimate the behavior of the system under different conditions.

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