

Experimental investigation, modeling, and optimization of combined electro-(Fenton/coagulation/flotation) process

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Original Article

Abstract

In this study, a combined electro-(Fenton/coagulation/flotation) (EF/EC/EI) process was studied via degradation of Disperse Orange 25 (DO25) organic dye as a case study. Influences of seven operational parameters on the dye removal efficiency (DR%) were measured: initial pH of the solution (pH₀), applied voltage between the anode and cathode (V), initial ferrous ion concentration (C_{Fe}), initial hydrogen peroxide concentration (CH₂O₂), initial DO25 concentration (C_O), applied aeration flow rate (FAir), and process time (tP). Combined design of experiments (DOE) was applied, and experiments were conducted in accordance with the design. The experimental data were collected in a hand-made laboratory-scale glass cylindrical batch reactor equipped with four graphite bar cathodes, an aluminum sheet anode, an aeration pump equipped with an air filter and air distributor, a 150-rpm mixer, and a DC power supply. A DR% of 98 was achieved with a pH₀ of 4, V of 10, C_{Fe} of 7.5, CH₂O₂ of 0, C_O of 140, and FAir of 0. The data were used for modeling using normal and reduced multiple regression models (MLR & r-MLR) and artificial neural networks (ANN & r-ANN). Further statistical tests were applied to determine the models' goodness and to compare the models. Based on statistical comparison, ANN models clearly outperformed the stepwise multiple linear regression (SMLR) models. Finally, an optimization process was carried out using a genetic algorithm (GA) over the outperformed ANN model. The optimization procedure was used to determine the optimal operating conditions of the combined process.

KEYWORDS: Fenton Reagents Concentration, Artificial Neural Network, Genetic Algorithm, Dye Removal Efficiency, Electro-(Fenton/Coagulation/Flotation)

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Introduction

Several methods have been developed for the removal of organic pollutants to decrease their impact on the environment, including adsorption,¹⁻⁴ photocatalysis,⁵ oxidation processes,⁶ and microbiological degradation.⁷ The application of techniques,⁸ electro-flotation,⁹ Fenton,¹⁰ electro-Fenton,¹¹

coagulation, and electro-coagulation processes,¹² as well as some combinations of these methods has also been assessed in recent years. The previous reports revealed that the efficiency of pollutant removal depends on the method and operational parameters.^{6,13,14}

The difficulties in degradation of new resistant organic compounds have inspired research to use combined procedures to increase efficiency and reduce process time by using synergetic effects. But there are

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drawbacks of the complex design of a combined reactor and the interference of coupled methods in a common reactor. Finding the proper combination of methods with high compatibility is a new research interest.

The electrocoagulation (EC), electro-Fenton (EF), and electroflotation (EL) techniques are considered to be potentially effective approaches to treat several types of wastewaters.^{15,16} EC consists of metallic hydroxide flocs generation within the wastewater by electro-dissolution of soluble anodes, which are usually made of iron or aluminum. The hydrogen gas produced at the cathode in the electrocoagulation unit could create and float the flocs. In other words, EC mainly plays a role in destabilizing and aggregating fine particles, and EL is responsible for floating the flocs formed in the effluent of EC unit. It makes it easy to use the synergetic effect of both EC and EL. In addition, because EC happens in the anode and floatation happens in the cathode, there is no interference between the processes. The several publications about EC and EF prove the compatibility of the combined methods.¹⁷

The oxidation of alcohols in the presence of H_2O_2 and $\text{Fe}(\text{H}_2\text{O})_6^{2+}$ was reported by Fenton in 1894.¹⁸ Among the advanced oxidation processes (AOPs), Fenton's treatment was found to be effective for treating wastewater containing several organic pollutants such as phenols, aniline, herbicides, and dyes. Fenton's treatment has two different stages. First, Fenton's oxidation occurs based on hydroxyl radicals ($^{\circ}\text{OH}$), and second, Fenton's coagulation occurs based on ferric coagulation following the oxidation stage. In situ generation of Fenton reagents induced by electrochemistry has become the focus of increasing research because it can oxidize organic compounds quickly and economically. Many persistent organic pollutants have successfully been degraded by the method. This process consists of electrogeneration of H_2O_2 by the reduction of dissolved oxygen on the electrodes such as

graphite, a mercury pool, carbon fiber, carbon-felt, carbon-polytetrafluoroethylene, and generation of Fe^{2+} by the reduction of externally applied Fe^{3+} . This avoids the high cost of H_2O_2 , maintains an almost constant H_2O_2 concentration, and regenerates Fe^{2+} more effectively.¹⁹

It is important to see that H_2O_2 and Fe^{2+} production and regeneration accrue in the cathode. It makes it easy to merge the process with electrocoagulation that happens in the anode electrode. In addition, at the proper current intensity and voltage, production of H_2 from the reduction of water continues in the cathode. Then, the combined method of EC/EF/EL has a synergistic effect with no interference of processes. Although combinations of both EC/EF and EC/EL have been used and studied in water and wastewater treatment, the efficiency and effective empirical parameters of the EC/EF/EL combination have not been studied in previous research. It is important to examine the feasibility of the EC/EF/EL combination and to investigate the effects of operational variables on its performance.

The first objective of this study was to report the degradation of Disperse Orange 25 (DO25) organic dye as a case study. This was done through a synthetic aqueous solution using a simple undivided electrochemical cell equipped with an aluminium (Al) anode for electrochemical generation of Al^{3+} and a graphite cathode for electrochemical regeneration of Fe^{2+} and H_2O_2 and H_2 production. The second objective of the study was to investigate the effects of seven operational parameters, including initial pH of the solution (pH0), applied voltage between the anode and cathode (V), initial ferrous ion concentration (CFe), initial hydrogen peroxide concentration (CH_2O_2), initial DO25 concentration (C0), applied aeration flow rate (FAir), and process time (tP). The effects of these parameters on the dye removal efficiency (DR%) were measured using a combined process by means of design of experiments (DOE). The

last objective was the application and assessment of an artificial neural network (ANN) and genetic algorithm (GA) for process modeling and optimization.^{13,14,20}

Materials and Methods

Ferrous ammonium sulfate was used to prepare a ferrous standard solution. Thirty percent hydrogen peroxide solution (Merck, Germany) was used for stock solution preparation by dilution. The hydrogen peroxide stock solutions were standardized using a titration method with a permanganate reagent solution (Merck, Germany) that was standardized by using a standard oxalic acid reagent solution (Merck, Germany) and a titration method. DO25 (C₁₇H₁₇N₅O₂) was purchased from AlvanSabet Co., Iran.

Stock solutions of synthetic wastewater were prepared by dissolving a desired amount of DO25 powder in distilled water. The desired concentrations of DO25 were prepared by diluting the stock solution. Graphite electrodes were purchased from KIG-Co., Iran. The characterization of the graphite electrodes is presented in table 1. Aluminum sheet electrodes were purchased from a local seller and prepared by cutting to a desired size. pH0 of the solutions was adjusted using NaOH (1 M) and H₂SO₄ (1 M) (Merck, Germany).

Statistical design of experiments is a useful technique for investigating a phenomenon by performing a minimum number of experiments. In this study, a 32-run Taguchi design of experiments was applied for investigation of pH0, V, CFe, CH₂O₂, C0, FAir, and tP using Minitab 14. The details of the 32 designed runs are presented in table 2. Furthermore, tP was investigated in eight levels for each run. The

levels of each parameter and its interval were determined by pre-tests.^{20,21}

The handmade reactor consisted of an 850-ml cylindrical glass reactor (9.5 cm diameter, 12 cm height), a 120-rpm magnetic stirrer, a DC power supply (RXN-303D-II, Zhaoxin Electronic Tech. Co.), an aluminum anode, and four connected graphite cathodes. The anode was made of aluminum sheets (40 × 100 × 1 mm) with an effective immersed surface area of 40 cm². Each cathode was made of a graphite bar [10 mm (diameter) × 120 mm (height)] with an effective immersed surface area of 3 cm². A cubic arrangement of four electrodes was used to increase the effective current intensity. The electrodes were placed vertically and dipped in 500-ml synthetic waste solutions. The distance between electrodes was fixed at 1 cm (Figure 1).

In each run, a 500-ml DO25 solution was decanted into the reactor. The empirical parameters were adjusted to the desired value based on design of experiments. In each eight desired tP, a 10-ml sample was extracted at a specific position of the reactor using a sampling pipet. Samples were centrifuged for 3 minutes at 2000 rpm, and then DR% was calculated for the decanted solution. The concentration of samples was evaluated by a standard spectrophotometric method using a calibration curve and a DR5000 spectrophotometer. Because of matrix effects, λ_{max} was extracted experimentally from the zero-time sample spectra in each run. Finally, DR% was calculated for samples using equation 1:

$$DR\% = (1 - C/C_0) \times 100 \quad (1)$$

where C₀ and C are the concentration of the solution before and after the process, respectively.

Table 1. The characterization of graphite electrodes.

Characteristic	Characteristic	Characteristic	Characteristic
Impregnation	None	Density (g/cm ³)	1.70
Flexural strength (mpa)	55	Compressive strength (mpa)	155
Modulus	22.000	Rockwell ball size/load	5/100
Hardness	105	Thermal conductivity (w/mk)	12
Thermal expansion (10 ⁻⁶ /k)	3		

The presented characterization was certified by the KIG. Co.

Table 2. The parameter levels for 32 runs.

Run	pH ₀	V (v)	C _{Fe} (ppm)	C _{H₂O₂} (ppm)	C ₀ (ppm)	F _{Air} (l/min)
1	2	0	0	0	20	0
2	4	5	2.5	1.5	20	0
3	6	10	5.0	2.7	20	0
4	8	15	7.5	4.3	20	0
5	2	5	2.5	0	80	0
6	4	0	0	1.5	80	0
7	6	15	7.5	4.2	80	0
8	8	10	5.0	1.6	80	0
9	2	15	5.0	5.0	140	0
10	4	10	7.5	0	140	0
11	6	5	0	6.1	140	0
12	8	0	2.5	4.5	140	0
13	2	10	7.5	2.0	200	0
14	4	15	5.0	0	200	0
15	6	0	2.5	6.1	200	0
16	8	5	0	4.1	200	0
17	2	15	0	6.1	20	2
18	4	10	2.5	4.1	20	2
19	6	5	5.0	2.0	20	2
20	8	0	7.5	0	20	2
21	2	10	2.5	6.2	80	2
22	4	15	0	4.5	80	2
23	6	0	7.5	2.3	80	2
24	8	5	5.0	0	80	2
25	2	0	5.0	4.2	140	2
26	4	5	7.5	7.0	140	2
27	6	10	0	0	140	2
28	8	15	2.5	2.3	140	2
29	2	5	7.5	4.1	200	2
30	4	0	5.0	6.7	200	2
31	6	15	2.5	0	200	2
32	8	10	0	2.0	200	2

V: applied voltage between the anode and cathode; C_{Fe}: initial ferrous ion concentration; C_{H₂O₂}: hydrogen peroxide concentration; C₀: initial DO25 concentration; F_{Air}: applied aeration flow rate

Results and Discussion

In all 32 runs, DR% of the samples at different tP was determined. The obtained DR% is presented in the four diagrams of figure 2. As can be seen from the diagrams, different operational parameters caused DR% to change from less than 1 up to 98 under experimental conditions of pH₀ of 4, V of 10, C_{Fe} of 7.5, C_{H₂O₂} of 0, C₀ of 140, and F_{Air} of 0. The combined method could be very effective to degrade DO71 when the operational parameters were fixed at the optimum conditions. Based on the results of run 7 presented in figure 2, a DR% of 90 was achievable in less than 20 minutes. This shows that the combined method could degrade the sample dye more rapidly than previously presented methods. In addition, it is clear evidence of the compatibility of the methods that were combined.

Some data were removed, and the 230 data that remained are presented in figure 2. The diagrams illustrate the influences of operational parameters on DR%.

Opposite to tP, which had obvious positive influences on DR% that were reported frequently, the influences of the six other parameters were unclear (Figure 1). It is essential to use statistical tools to clarify the parameters' influences and to distinguish the effective parameters for modeling and optimization.

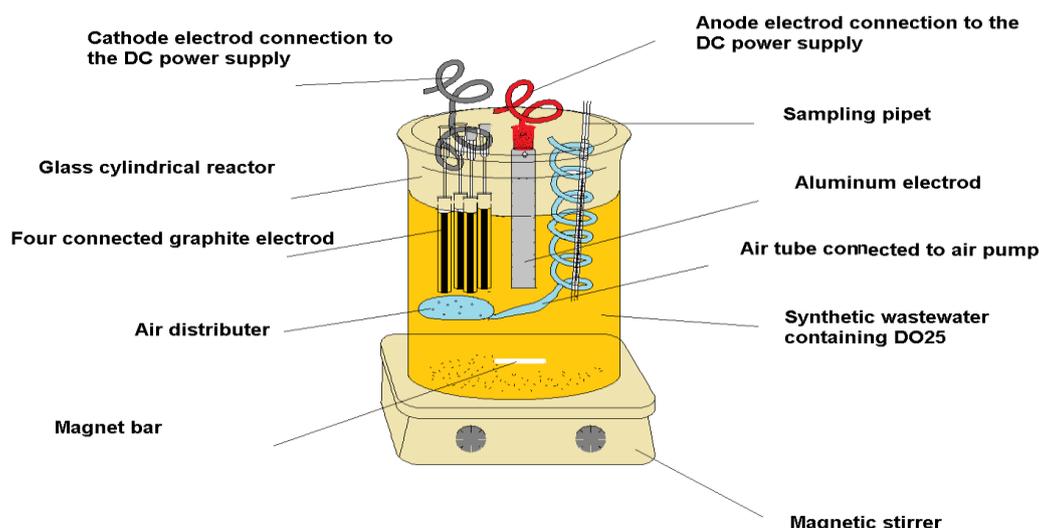


Figure 1. The lab-scale batch experimental setup of the process unit

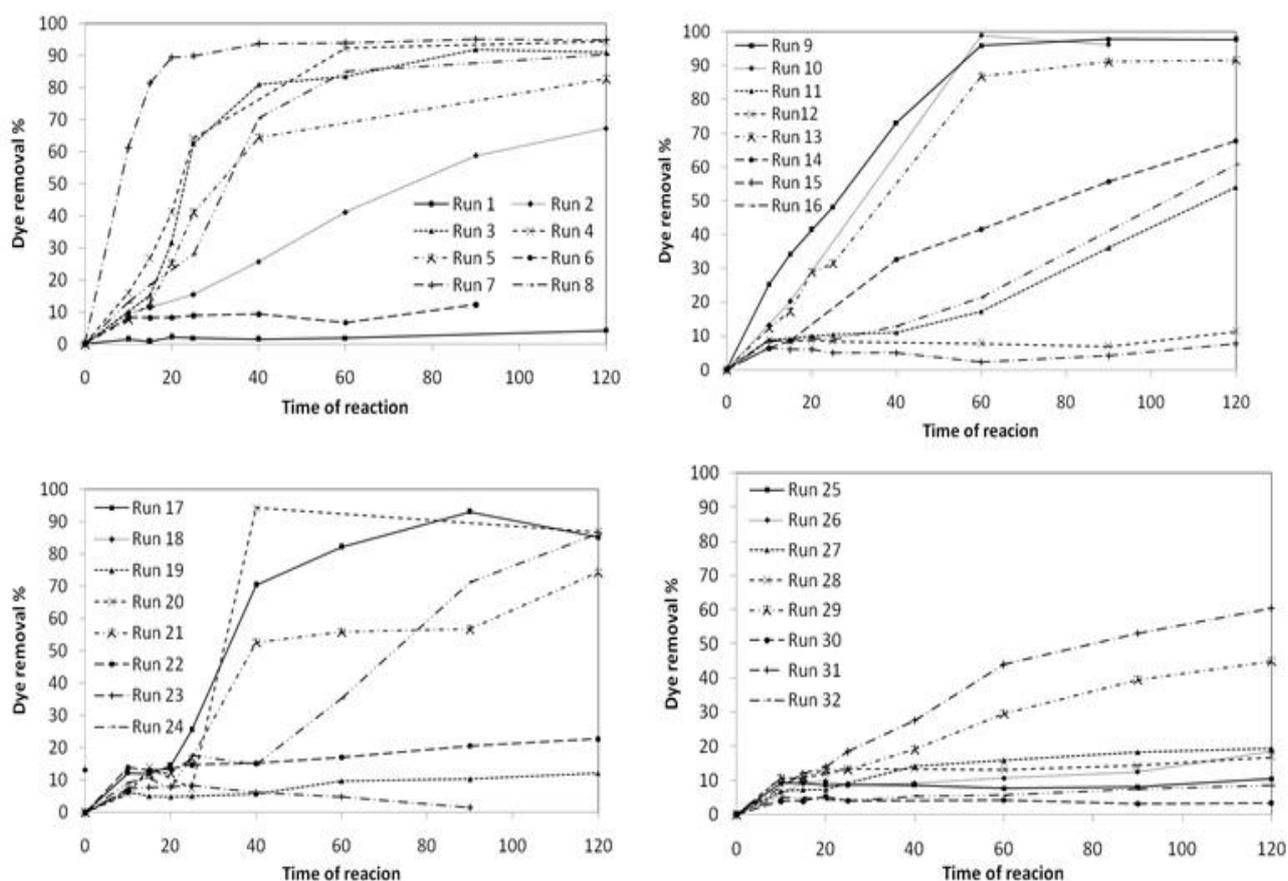


Figure 2. The experimental dye removal efficiency (DR%) results of 32 runs

Multiple linear regression (MLR) and stepwise multiple linear regression (SMLR) were then applied to investigate the parameters' influences. The MLR model was developed for DR%, and the statistical details of the model are shown in table 3.

Based on the coefficients presented in

table 3, among the parameters, t_P , V , CH_2O_2 , and C_{Fe} had positive influences, which has been reported frequently and is logical too. F_{Air} , C_0 , and pH had negative influences on DR%. The negative influences of pH and C_0 are frequently reported in this kind of study, when the negative effect of air should be

Table 3. The multiple linear regression (MLR) and reduced multiple linear regression (r-MLR) models along with statistics of parameter influences

Parameters	MLR				r-MLR			
	coeff.	St. coeff.	t-value	P	coeff.	St. coeff.	t-value	P
constant	1.36	-	0.29	0.77	-2.40	-	-0.68	0.50
pH_0	-0.90	-0.07	1.74	0.08	-	-	-	-
V	2.04	0.32	-1.85	0.06	2.04	0.39	9.77	< 0.01
C_{Fe}	2.79	0.23	8.94	< 0.01	2.81	0.27	6.70	< 0.01
$C_{H_2O_2}$	0.39	0.01	6.45	< 0.01	-	-	-	-
C_0	-0.06	-0.15	-0.31	0.75	-0.06	-0.14	-3.54	< 0.01
F_{Air}	-8.84	-0.28	-4.12	< 0.01	-8.47	-0.29	-7.26	< 0.01
t_P	0.39	0.55	-7.64	< 0.01	0.39	0.51	12.76	< 0.01
Model goodness parameters	MLR				r-MLR			
	F value	R^2	P	RMSE	F value	R^2	P	RMSE
	62.80	57.10	< 0.01	18.00	73.70	0.59	< 0.01	19.00

MLR: Multiple linear regression; r-MLR: Reduced multiple linear regression; RMSE: Root-mean-square error; Coeff: Coefficient; St. coeff: Standard coefficient

interpreted. It is important to see that based on the unbiased standardized coefficient presented in table 3, CH_2O_2 and pH almost had a negligible effect. However, the negligible influences for H_2O_2 and especially for pH are questionable, but it may accrue due to conflict of the combined methods and compensation of the negative and positive influences. Based on the unbiased parameters, tp, V, and FAir had more important influences on DR%.

The statistical parameters obtained from analysis of variances (ANOVA) showed that influences of some parameters were meaningless and should be omitted from the model. Stepwise multiple linear regressions were used to remove the meaningless variables as a common appropriate statistical tool. The reduced MLR model (r-MLR) obtained using the stepwise algorithm is presented in table 3 along with its statistical details. As presented in table 3, two parameters, pH and C0, were removed by the SMLR algorithm in a significant alpha level of 0.05.

Furthermore, table 3 and the model goodness parameters indicate that the MLR and r-MLR models did not have good predictability for DR%. This can arise from the complex mechanism of the process. The simple linear method of MLR and SMLR could not model the process and could not distinguish the nonlinear influences or interaction of the important parameters such as pH and C0. The artificial neural network (ANN) is a parallel computational procedure consisting of highly interconnected processing element groups called neurons. Owing to their inherent nature to model and learn "complexities," ANNs have found wide applications in various areas of wastewater treatment. This has inspired us to use ANN as a more powerful nonlinear modeling approach to obtain a good predictable model. Besides, the ANN was applied to determine the importance of pH and C0 in a nonlinear ANN model. A reduced ANN (r-ANN) model and ANN model based on r-MLR and MLR model parameters were constructed.

Therefore, the seven and five operational parameters were applied as inputs of the ANN and r-ANN models, respectively, while DR% was considered as the dependent variable. The data set was randomly divided into three parts: 60% (157 data) as a training set, 20% (53 data) as a validation set, and 20% (52 data) as a testing set. The training set was used to adjust the parameters of the models, the testing set was used to calculate its estimation power, and the validation set was used to prevent over-training. A back propagation algorithm has been used because it is very fast and can be employed quite easily. The number of hidden layers, the neurons of each hidden layer, and the learning rate were determined via trial and error.

The best selected net had one hidden layer with 11 neurons and a learning rate of 0.21 (7:11:1 net) for the ANN model. The best selected net for the r-ANN model had one hidden layer with 8 neurons and a learning rate of 0.17 (5:8:1 net). The "tansig" transfer function was selected for the input and hidden layers, and "purelin" was selected for the output in both models. Once the networks were trained, the weights and bias of each neuron and layer were saved in the ANN model. Then, they were used to estimate the test set. Finally, the consistency of the ANN models was revealed by tests quantified with predictive Q^2 and R^2 , while the reliability or accuracy of the models was revealed by root-mean-square error (RMSE). The test results of the ANN and r-ANN model goodness are presented in table 4.

The comparison of table 3 and table 4 clearly confirms that the ANN models outperformed the MLR models. This confirms the complex nonlinear nature of the process. In addition, the significant decline in goodness of the r-ANN rather than ANN not only confirms the importance of pH and C dye, but also proves the nonlinear complex nature of the process. As a goal of each modeling study, optimization of the process was considered. The optimized parameters can be used to identify the quality and quality of parameter influences.

Table 4. Statistical characteristics of the artificial neural network (ANN) model

Model	ANN			r-ANN		
	Train set	Validation set	Test set	Train set	Validation set	Test set
R ²	0.99	0.97	0.96	0.98	0.92	0.91
Q ²	0.99	0.97	0.96	0.98	0.92	0.91
RMSE	2.10	6.20	5.10	3.50	9.80	7.90

ANN: Artificial neural network; r-ANN: Reduced artificial neural network; RMSE: Root-mean-square error

A genetic algorithm (GA) was used to optimize the experimental parameters using the best obtained models. A GA is an adaptive heuristic search algorithm based on the evolutionary ideas of natural selection and genetics.

The GA toolbox in MATLAB software was used for generating the optimal solution for DR% using the "ga" function. Four MATLAB functions using four MLR, r-MLR, ANN, and r-ANN models were written for creating fitness functions for the optimization problem. The DR% component to be maximized was negated in the vector valued fitness function since "ga" minimizes all the objectives. The result of the GA solution is shown in table 5.

As can be seen from table 5, the optimization process proved that the ANN and r-ANN models were more compatible with the experimental results predicting almost 99% as an optimum value of DR%. In addition, the obtained optimum values of experimental parameters were different for these four models. The ANN and r-ANN models were more powerful and accurate and should have more logical values.

In this study, the effect of initial pH was investigated in the range of pH 2 to 8. The optimum pH values were 6.1 in the ANN model and 2.1 in MLR. Both pH 2.1 and 6.1 are acidic conditions that are frequently

reported as more desirable conditions for this kind of process, but there are meaningful differences between pH 2.1 and 6.1 that can be judged by the ANN and MLR model quality. However, the low value of pH 2.1 causes more hydrogen electrogeneration or Fenton reagent efficiency, but pH 6 is in an efficient range for electrocoagulation using aluminum. It may be logical that a combination of several methods has several optimum values for one parameter like pH.

The optimum values for voltage ranged from 0.3 to 15 V. However, more voltage makes the electrodic process easy, but it can cause inversion of the efficiency in the electrocoagulation process by producing more coagulant than the efficient amount. Regarding the Fenton reagents, Fe²⁺ and H₂O₂, the maximum available concentrations in their investigated ranges (7.5 ppm) was selected as the optimum values by four optimization processes. This arises from the importance and strength of the Fenton and electro-Fenton methods compared to other methods. In addition, it may rise from bad range selection for these parameters.

Table 5 shows that higher C₀ and lower A made it essential to apply more t. It is frequently reported that higher C₀ necessitates more time for the process. In addition, less aeration makes the flotation and electro-Fenton processes weak.

Table 5. Genetic algorithm optimization result

Model	Parameters							
	pH ₀	V (volt)	D _{Fe} (ppm)	D _H (ppm)	C ₀ (ppm)	A	t	DR%
ANN	6.1	0.3	3.1	7.2	22	0.5	8	98.9
r-ANN	-	11	6.7	-	130	2.0	20	96.5
MLR	2.1	15	7.5	7.4	21	0.0	10	56.0
r-MLR	-	15	7.4	-	20	0.0	16	53.3

ANN: Artificial neural network; r-ANN: Reduced artificial neural network; MLR: Multiple linear regression; r-MLR: Reduced multiple linear regression; V: applied voltage between the anode and cathode; D_{Fe}: Fe ion concentration; C₀: initial DO₂₅ concentration; A: Aeration flow rate; t: ; DR: Dye removal efficiency;

Conclusion

The present study clearly showed the power of compatible combined methods of EF/EC/EI as a fast and applicable method for degradation of DO25. The study also confirmed the deep influences of several operational parameters on the efficiency of the method. The applied design of experiment, modeling, and optimization processes were successful for obtaining the determined statistical goals. The artificial intelligence systems such as ANN and GA that were applied in the study presented acceptable performance in the modeling and optimization of the complex combined method. The statistical tools were applied to distinguish the effective operational parameters and also to determine the quantity of the influences.

Conflict of Interests

Authors have no conflict of interests.

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