Hindawi
International Journal of Chemical Engineering
Volume 2022, Article ID 1812776, 11 pages
https://doi.org/10.1155/2022/1812776

Research Article

Using Sono-Electro-Persulfate Process for Atenolol Removal from Aqueous Solutions: Prediction and Optimization with the ANFIS Model and Genetic Algorithm

Nasrin Zahedi,[1,2] **Bahare Dehdashti**,[1,2,3] **Farzaneh Mohammadi**,[1] **Maryam Razaghi**,[1,2] **Zeynab Moradmand**,[1,2] **and Mohammad Mehdi Amin**[1,3]

[1]*Department of Environmental Health Engineering, School of Health, Isfahan University of Medical Sciences, Isfahan, Iran*
[2]*Student Research Committee, School of Health, Isfahan University of Medical Sciences, Isfahan, Iran*
[3]*Environment Research Center, Research Institute for Primordial Prevention of Non-communicable Disease, Isfahan University of Medical Sciences, Isfahan, Iran*

Correspondence should be addressed to Bahare Dehdashti; baharehdehdashty@yahoo.com and Mohammad Mehdi Amin; mohammadmehdia@gmail.com

Received 9 September 2022; Revised 11 October 2022; Accepted 20 October 2022; Published 4 November 2022

Academic Editor: Ho SoonMin

Atenolol (ATN) is a drug that is widely used to treat some heart diseases, and since it cannot be completely decomposed in the human body, some amounts of it are found in surface water. These amounts may bring risks to the environment and humans, and for this reason, its removal is a must. In the present study, the combined sono-electro-persulfate method was used for ATN removal. Based on the design of the experiment conducted by response surface methodology (RSM), the effects of 5 main factors (pH, time, PS concentration, current intensity, and initial ATN concentration) have been investigated at 5 levels. After passing the test steps in different conditions, the remaining amount of ATN has been measured by high-performance liquid chromatography (HPLC). Finally, an adaptive neuro-fuzzy inference system (ANFIS) with 99.63% accuracy and a genetic algorithm (GA) were used to analyze and interpret data and predict optimal conditions. The obtained results indicate the possibility of a maximum efficiency of 99.8% in the mentioned conditions (Ph of 7.4, time of 18 min, PS concentration of 2000 mg/L, current intensity of 3.35 A, and initial ATN concentration of 11.2 mg/L). According to the obtained results, the initial concentration of ATN can be considered as the most effective factor in this process, and the best Ph range for this experiment was the neutral range. The sono-electro persulfate process can be mentioned as a new and effective method for removing ATN from water sources.

1. Introduction

In recent decades, the use of pharmaceutical and personal care products (PPCPs) has greatly expanded [1, 2]. Although these products help to improve the quality of life, their existence in the environment and water resources can have very harmful effects [1].

Drug contamination of water sources can be caused in several ways. The most important of which include domestic, hospital, and veterinary waste waters [2]. Since the human body does not have the ability to fully metabolize beta-blocker drugs, some amounts of these drugs enter surface water and finally drinking water sources [3]. Consumption of this polluted water may have risks for human health, such as disturbance in testosterone levels, reduction of fertility, and behavioral disorders due to its consumption by aquatic animals [4]. Atenolol (4-(2-hydroxy-isopropylaminopropoxy)-phenylacetamide) [5] is widely used to treat disorders such as high blood pressure, chest pain, heart disease [6], and for the treatment or prevention of heart attacks [5]. In some cases, beta-blockers (ATN) may have adverse effects on nontarget tissues, such as affecting the endocrine glands and thus disrupting testosterone levels in men [7]. Therefore, the removal of ATN from aqueous solutions is very important due to its potential danger [6].

Methods of removing PPCPs from water sources include simple to advanced treatment processes, such as membrane processes, adsorption, coagulation, activated sludge, advanced oxidation, etc. [8].

Among the common water and wastewater treatment processes, advanced oxidation processes (AOPs) have received much attention due to the conversion of toxic pollutants into less toxic substances. Moreover, this remarkable performance in the removal of organic pollutants has attracted enormous interest [9, 10].

AOPs are the methods in which organic matter is oxidized or converted to minerals by producing free radicals such as hydroxyl [11]. The evolution of information in 2000 led to the promotion of the AOPs method based on sulfate radicals [10]. To produce sulfate radicals, suitable oxidants such as peroxymonosulfate (PMS) and persulfate (PS) must first be activated [10].

There are several ways to activate these oxidants and produce radicals such as ultraviolet (UV), ultrasonic (US), microwave, visible light, electrolysis, heat, or a combination of them [9, 10].

The advanced oxidation process with electrolysis (E-AOP) is one of the newest nonthermal methods for water and wastewater treatment [12], which seems to have received much attention due to its high efficiency in the conversion of organic compounds [13]. The advantages of this method include low operating costs, simple equipment, relatively low capital, flexibility [14], high efficiency, low toxicity of reactants [15], and the result of very low environmental pollution [14].

The use of ultrasonic waves for purification causes the decomposition of pollutants and the creation of sound holes in water (in the form of small bubbles) [15], which, when energy is concentrated in these holes, creates free radicals [13]. Ultrasonic waves are very useful for many applications, such as cellular disorders, emulsions, nanotechnology, wastewater treatment, etc. Its main advantages are high safety, high water penetration, high decomposition efficiency, and low energy requirements [16].

If the advanced electrochemical oxidation process and ultrasonic waves are used in combination to generate free radicals, we will see positive effects such as [15] an increase in electrochemical efficiency and overall process efficiency, prevention of electrode deposition [17], an increase in reaction speed [13], and more compatibility with the environment. One of the most important methods of forecasting and optimization using statistical and mathematical algorithms is the response surface method (RSM), artificial neural network (ANN), and genetic algorithm (GA) [18].

ANFIS is a modern and effective approach to modeling input-output relationships in complex systems [19]. In this method, it is possible to learn from training data as an ANN and then solve problems on a fuzzy inference system (FIS). Finally, the hidden layers are identified exactly by an FIS in the ANFIS network. This method eliminates the important challenge of determining and predicting the hidden layers in the ANN model. This can be a strong reason for using the ANFIS method while this approach does not have a complex mathematical model and is a fast and flexible method for developing predictive models of chemical treatment processes [20].

GA is a research method to obtain accurate or approximate solutions to optimization problems [20]. GAs are optimization techniques that are controlled based on the principles of evolution and natural genetics [21]. One of the advantages of this method is the ability to create clear models for complex and difficult systems. As a result, we can use this method for problems that for various reasons, such as discontinuous, random, and nonlinear functions, are not suitable for standard optimization patterns [20].

Considering the negative effects of ATN remaining in surface water and the ineffectiveness of simple and basic water purification methods in removing this substance, in this study we aimed to achieve the main goal of removing or reducing it effectively. The aim of this study was to use the sono-electro-persulfate, which is a new and environmentally friendly method, to investigate the efficiency of the process for degradation of ATN from aqueous solutions by the response surface method. Moreover, for the prediction and optimization of experimental data, ANFIS and GA algorithms were used, respectively. Also, in order to investigate the trend of changes in the concentration of ATN over time, the kinetics of the reaction were investigated.

2. Materials and Methods

2.1. Materials. Atenolol, known by its chemical name as 2-(4-(2-hydroxy-3-(propan-2-ylamino) propoxy) phenyl) acetamide, with reported purities of >98% was obtained from Raha Pharmaceutical Factory (Isfahan, Iran). Also, ammonium persulfate ($S_2O_8NH_4$), hydrochloric acid (HCL, 0.1 M), sodium hydroxide (NaOH, 0.1 M), potassium dihydrogen phosphate (PDP), humic acids (HA), HPLC-grade acetonitrile, and methanol (for rinsing injection needles) were purchased from Merck & Co., Germany.

To prepare an ATN stock solution, 1 g of ATN powder was added to 1 L of deionized water and mixed well with a magnetic stirrer. An HA stock solution was prepared by adding 0.3 g of dry HA powder to 1 L of deionized water and stirring continuously for one night. The pH of the solutions was adjusted with 0.1 M HCl for pH < 6 and buffered with 0.1 M NaOH for $6 < pH < 12$. A digital ultrasonic bath (BANDELIN-DT255H) and a DC power supply (DAZHEN-PS 305D) were also used.

2.2. Reactor Setup and Experimental Procedure. In this study, a sono-chemical reaction chamber was used with a volume of 500 mL equipped with ultrasonic waves generated by an ultrasonic homogenizer and two iron electrodes (anode) and two copper electrodes (cathode). The active anode area was 36 cm^2 with a distance of 1 cm. The electrodes were in the form of floating inside the electric cell and connected to the direct current generator (Figure 1).

In this research study, five independent variables, including pH, reaction time, initial concentration of ATN, persulfate concentration, and electric current intensity, at five coded levels (−2, −1, 0, 1, and 2) were studied.

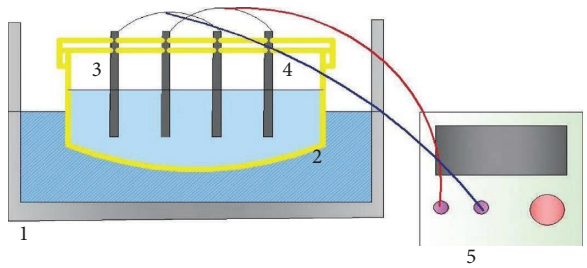


Figure 1: Schematic diagram of the reactor used in this study: (1) ultrasonic bath; (2) laboratory scale reactor; (3) iron electrode; (4) copper electrode; and (5) DC power supply.

After making the ATN stock solution (1 g/L), different concentrations of the drug based on the test design (1, 5, 10, 15, 20 mg/L) were used to examine the effect of the other parameters.

PS concentrations of 100, 250, 500, 1000, and 2000 mg/L were added to the solution. It was exposed to ultrasonic waves (35 kHz) and the intensity of variable electric currents (1.5, 2, 2.5, 3, and 3.5 mA). HCl and NaOH 0.1 M were used to apply different pH values (3, 5, 7, 9, and 11).

2.3. Design of Experiments. RSM is a method that, with the help of mathematical science and statistics, determines the optimal conditions for an experiment (VERY). RSM is an experimental statistical modeling method for multiple regression analysis using quantitative data [22].

In this study, for design of the experiments, the central composite design (CCD) method was used. An experimental design was performed in Design Expert 11.0 software. In this method, 5 levels were considered for each variable. The variables and their levels can be seen in Table 1.

2.4. Analysis Methods. High-performance liquid chromatography (HPLC-Jasco PU2080) with a UV detector (UV-2075) at 231 nm and a Waters-spheri-sorb-ODS2 column was used to achieve the residual ATN concentration in the experiment. Acetonitrile/potassium dihydrogen orthophosphate with a ratio of 70–30 was used as the mobile phase.

The ATN removal efficiency was calculated using equation (1)

$$R^2 = \frac{C_0 - Ce}{C_0} \times 100. \quad (1)$$

In equation (1), C_0 and C_e represent the initial concentration and the equilibrium concentration, respectively.

2.5. ANFIS Model. The ANFIS model is based on the Sugeno structure with a fuzzy neural designer created in MATLAB 2017 software. The ANFIS architecture is a combination of both artificial neural networks and fuzzy logic, and the mapping relationship between input and output data defines the optimal distribution of membership functions.

Table 1: The variables and that levels in this study.

Factor	Name	Unit	Min	Max	Levels
A	ATN	mg/L	1.0	20.0	1, 5, 10, 15, 20
B	*pH*	—	3.0	11.0	3, 5, 7, 9, 11
C	Time	Min	5.0	60.0	5, 10, 20, 40, 60
D	Current density	*A*	1.5	3.5	1.5, 2, 2.5, 3, 3.5
E	PS	mg/L	100.0	2000.0	100, 250, 500, 1000, 2000

Basically, the five main layers of ANFIS model formation are as follows: (1) fuzzy layer, (2) product layer, (3) normalized layer, (4) non-fuzzy layer, and (5) total output layer.

In this study, several different membership functions such as triangular membership function (trimf), trapezoidal membership function (trapmf), generalized bell membership function (gbellmf), Gaussian curve membership function (gaussmf), and Gaussian hybrid membership function (gauss2mf) have been studied. P-shape membership function (pimf), difference between two sigmoid membership functions (dsigmf), and product of two sigmoid membership functions (psigmf) are all examples of membership functions.

In this study, to identify the most appropriate membership function, the data set obtained (32 samples) from AOP (sono-electro-persulfate process) was randomly divided into two categories: 80% training and 20% testing. In order to increase the convergence rate, a hybrid learning algorithm was used in this research. This algorithm was used to integrate the two methods of least squares and gradients with the aim of updating the introduced parameters. The least squares method was used to optimize the next parameters. After determining the optimal subsequent parameters, the descent gradient method was used to better adjust the default parameters. The ANFIS output was determined by the following parameters [23].

The accuracy and adequacy of the models presented by ANFIS were determined by the determination coefficient (R^2) and root mean square error (*RMSE*) (equations (2) and (3)).

$$R^2 = \frac{1}{N} \frac{\sum\left(Y_{pr\,d,i} - Y_{pr\,d,m}\right)\left(Y_{\exp,i} - Y_{\exp,m}\right)}{\sigma_{pr\,d}\sigma_{\exp}}, \quad (2)$$

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(y_{pr\,d,i} - y_{\exp,i}\right)^2}. \quad (3)$$

2.6. GA Optimization. The GA was implemented in the MATLAB software R2017a version. GA was used to obtain the optimal conditions for ATN removal using the ANFIS model developed in this study as fitness function as follows:

Fitness function = Max (Removal rate from ANFIS simulation).

Other settings intended for genetic algorithms were number of generations 250, the rank scaling function, the selection function of Stochastic uniform, the number of elite 2, the crossover fraction equal to 0.8, and the mutation function of constraint dependent and combination of scattered functions [20, 24].

2.7. Scavengers. The effect of confounders, including chlorine, nitrate, and humic acid, on the removal reaction of ATN, at the optimal test conditions, was investigated using values of 0, 10, 100, and 500 mmol for each of the confounders.

The interference effect of organic matters was examined using humic acid. The stock solution of humic acids was made by mixing 0.3 g of dry humic acid powder with 1 liter of distilled water. It was stirred overnight at 100 rpm. The solution was then filtered through a 0.7 μm fiberglass filter. The UV absorption at 254 nm (UV-254) was used for analysis of the humic acid content. Chloride and nitrate concentrations were also measured [25].

2.8. Kinetics of the Model. In this study, after finding the highest efficiency, among the examined steps, four parameters (ATN, pH, current density, and PS) were kept constant, and only the variable of time was changed as 4, 8, 12, 16, and 20 min.

3. Results and Discussion

3.1. Modeling with ANFIS. The ANFIS structure and network model were used for this work to predict ATN removal. The results are shown in Figures 2(a) and 2(b).

The systems of Sugeno type FIS and membership function were used to do the training process. In this work, the FIS considers the 5 inputs of ATN initial concentration, pH, time, current density, and PS. In order to generate the modified rules for a certain data set, the network partitioning technique was used. 80% of the data set (26 samples) was used to train the ANFIS model and the remaining 20% of the dataset (6 samples) was used to test the predictive proficiency of the relevant model.

The ANFIS model first performs the training process for the training data set and then tests the results with the test data. In addition, in the ANFIS model training process, the input data set is drawn several times to prevent any possible errors. The required number of iterations in order to map is stated as epochs. It should be noted that the 20 epochs (i.e., iterations) are essential to accomplishing the training process on 26 data sets. Also, to measure the validity and efficiency of the model after the training process, this model was tested on 6 datasets.

In the next step, the results obtained from the training and testing processes of the ANFIS model were analyzed by using different types of membership functions. According to the obtained results, it can be said that the Gaussian membership function (gaussmf) has a lower prediction error than other functions. In fact, the RMSE is $2.509E-5$ for training and 1.607 for testing. As a result, it can be said that the ANFIS structure with optimal 2 2 2 2 2 membership performance is the best for this process. Figure 3(a) represents the training error curve for the experimental values of ATN removal. Figures 3(b) and 3(c) demonstrate the experimental results of ATN removal under similar processing conditions using the ANFIS model to compare training and experimental datasets. Moreover, R^2 in this study was about 99.63% and 99.24% for the training and testing data sets, respectively, which indicates the accuracy and adequacy of the model.

3.2. Optimization with GA. In the present study, the genetic algorithm was used to optimize the input parameters and finally to achieve the highest removal efficiency. The designed ANFIS model was introduced as a fitness function for GA. In Figure 4(a), the diagram of the best and average of fitness values in each generation is presented. Figure 4(b) also represents the best fitness values in the final generation. Therefore, the optimal values of the input parameters, which are ATN, pH, time, current density, and PS, were equal to 11.2 mg/L, 7.4, 18.0 min, 3.35 A, and 2000 mg/L, respectively, which led to the highest ATN removal efficiency of 99.80%.

3.3. Factors Affecting the Removal of ATN in the Sono-Electro-Persulfate System. Using the ANFIS model designed, the following diagrams were drawn in MATLAB software. The parameters shown inside each graph were considered variables, and the other parameters for each graph were fixed and equal to their optimal value (obtained from the genetic algorithm) (Figure 5).

3.3.1. Effect of Initial pH. pH is an important and determining factor in sono-electrolysis processes and it has a significant effect on energy consumption factors [13]. According to Figure 5(a), the pH has a nonlinear relationship with the removal efficiency. Rising the pH up to about 7 increases the removal efficiency but then decreases with increasing pH. These conditions have been observed in all studied concentrations. As can be seen, the highest removal efficiency was at $pH = 7$ and the lowest was at $pH = 3$.

When the water is acidic, the amount of dissolved oxygen in the water decreases, resulting in a decrease in the production of sulfate (SO_4^{2-}) ions. Given that sulfate ions are the main cause of oxidation. Reduction of sulfate ions reduces oxidation and thus reduces the removal efficiency of ATN from the aqueous medium. By changing the pH of the water and approaching the neutral state, the dissolution of oxygen in the water increases and consequently, the efficiency increases [9]. Due to the fact that an excessive increase in concentration has a negative effect on removal efficiency as expected, a decrease in efficiency at pH higher than 7 can be justified [26].

3.3.2. Effect of Initial ATN Concentration. Figure 5(a) shows that in this study, the removal efficiency increases with rising the concentration of ATN to 10 mg/L. Low removal

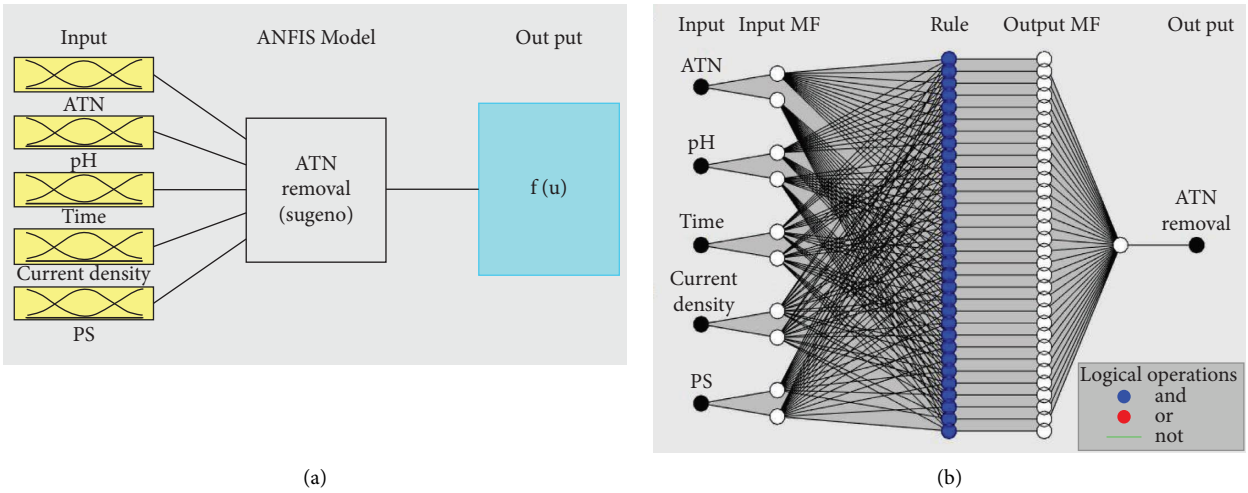


Figure 2: Developed (a) ANFIS structure. (b) Neural network model for ATN removal.

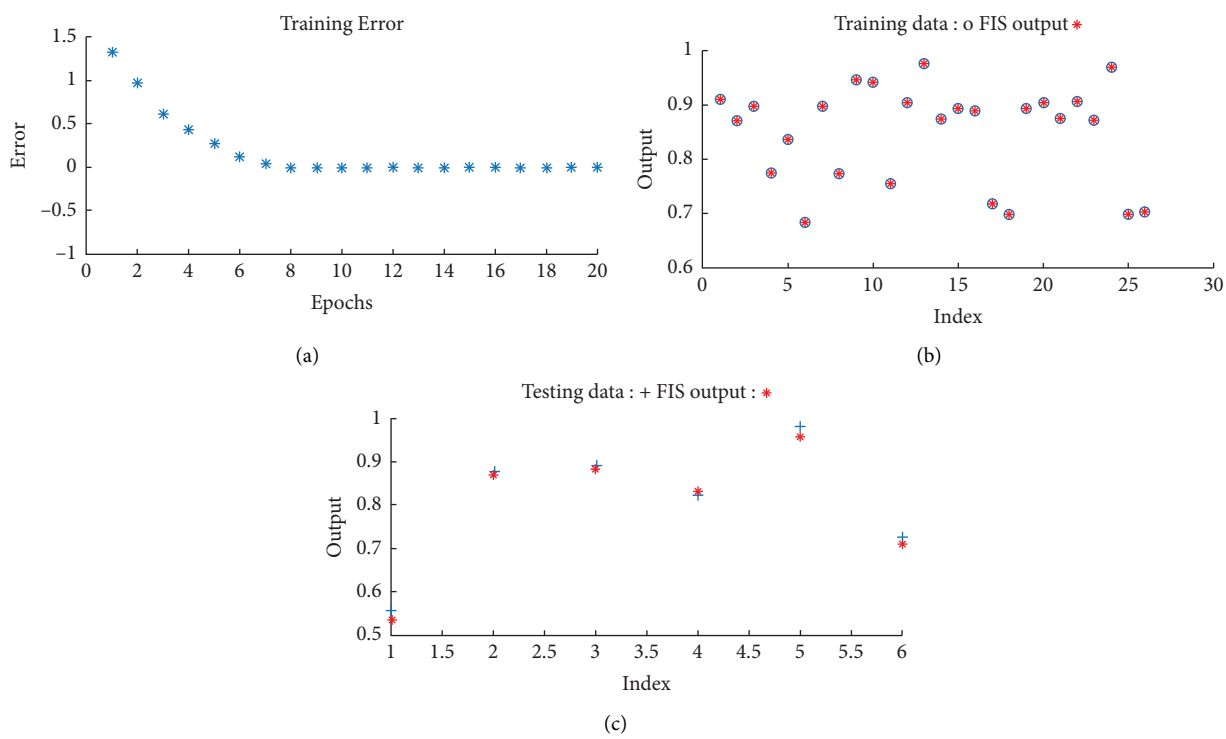


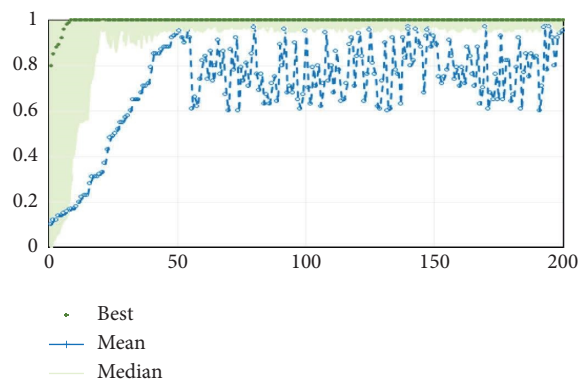
Figure 3: (a) Training error curve of the ANFIS model for ATN removal. (b) Comparison of experimental and predicted values in training. (c) Testing processes.

efficiencies at very low concentrations may be due to insufficient contact surface between atenolol molecules and ultrasonic waves.

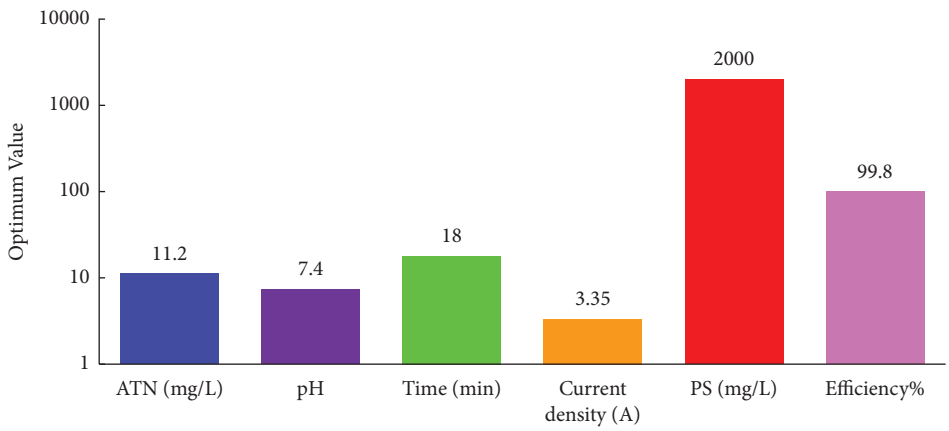
At ATN concentrations above 10 mg/L, the efficiency decreases with rising concentration. This decreasing trend may be due to the lack of a sufficient number of free radicals to react with the ATN molecules.

On the other hand, increasing the amount of intermediate products during oxidation reactions increases their competition with ATN for free radicals and thus reduces the efficiency [27].

3.3.3. Effect of Time. Figure 5(b) shows the variable of time versus the initial atenolol concentration and efficiency. With rising contact time, initially, the removal efficiency has increased with a steep slope, but after reaching the optimal time, which is about 20 minutes, the increase in efficiency has decreased with increasing time, and after that, the efficiency has remained constant. This may be due to the increased rate of consumption of sulfate ions in solution, followed by an increase in efficiency until the optimal time is reached [9].



(a)



(b)

Figure 4: Genetic algorithm results. (a) The best and average fitness values per generation. (b) The optimal value of the input parameters and highest removal efficiency.

3.3.4. Effect of Persulfate Concentration. In Figure 5(c), the persulfate variable had an almost nonlinear relationship with the removal efficiency, but its effect on the removal efficiency was less than the initial concentration. In any case, increasing the sulfate has improved the removal efficiency.

Since PS is the only source of SO_4^{2-} production in this oxidation system, increasing PS concentration increases the concentration of sulfate ions, which increases the efficiency of the system. It should be noted that some adverse reactions at high concentrations of sulfate reduce the removal efficiency (equations (4)–(7)) [28].

$$SO_4^- + SO_4^- \longrightarrow S_2O_8^{2-} \tag{4}$$

$$S_2O_8^{2-} + SO_4^- \longrightarrow S_2O_8^- + SO_4^{2-} \tag{5}$$

$$S_2O_8^{2-} + OH \longrightarrow S_2O_8^- + OH^- \tag{6}$$

$$SO_4^- + OH \longrightarrow HSO_4^- + \frac{1}{2}O_2 \tag{7}$$

3.3.5. Effect of Intensity of Electric Current. Figure 5(d) shows that the current density parameter has a linear relationship with the removal efficiency and its increase has led to an increase in the removal efficiency.

Current intensity is one of the parameters that is very effective in sono-electro-coagulation [13]. When the current intensity is low, the electrodes do not have enough power to oxidize [27].

According to Faraday's law, the intensity of the current is directly related to the solubility of the electrode and the charge created in the solution. Rising the current intensity increases the oxidation of the anode surface and the production of iron oxide ($Fe(OH)_3$). The presence of these oxides facilitates the deposition of contaminants and thus increases the removal efficiency [13].

Also, increasing the intensity of the current causes the bubbles to thicken and their size to decrease, which increases the level of absorption of pollutants [29]. In Table 2, the results of the current study are compared with some similar studies in terms of effectiveness in optimal conditions.

3.4. Sensitivity Analysis. A sensitivity analysis is performed to evaluate the significance of the effect of each parameter on the output variable [32]. This shows the extent to which each input factor affects the accuracy of the predicted model, and finally, it is a good way to design more applications [33].

In this study, sensitivity analysis was performed by the Pearson correlation method. Based on the following diagram, the effect of the input parameters was in the order of

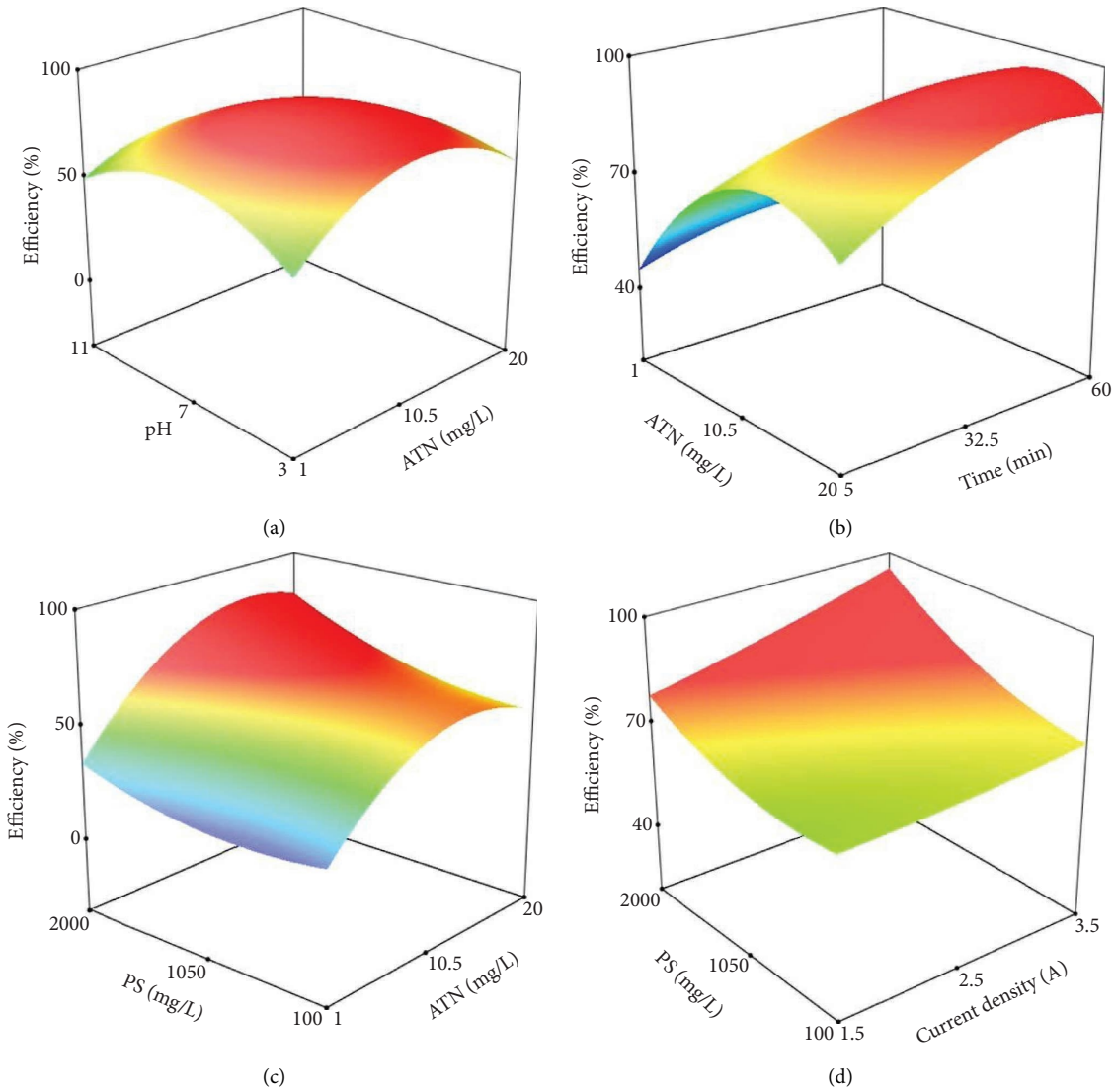


Figure 5: 3D graphs of ANFIS prediction showing the interaction effects of input variables on the ATN degradation rate.

Table 2: Comparative evaluation of the results for this study with previous studies.

Pollutant	Degradation method	*Optimum condition*					Efficiency (%)	References
		pH	Initial concentration of the pollutant (mg/L)	PS concentration	Time (min)	Current intensity (A)		
Cefixime	Sono-electro-fenton	3.07	10.4		81.5		97.50	[15]
Petrochemical wastewater	Sonoelectro-activated persulfate	3		20 (mM/L)	120		91.20	[9]
ATN	Oxidative treatments		10		30		94	[30]
ATN	By poly (AAM_MA) hydrogel	6			120		94.20	[31]
ATN	Sono-electro-persulfate process	7.4	11.2	200 (mg/L)	18	3.35	99.80	This study

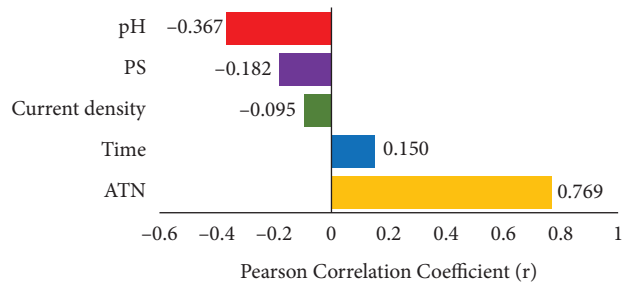


Figure 6: Sensitivity analysis using the Pearson correlation.

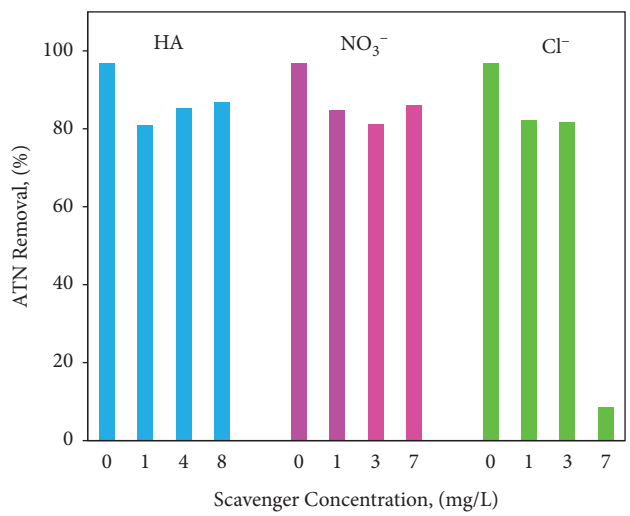


Figure 7: The effect of scavengers on the rate of atenolol removal.

Table 3: Kinetic equations for ATN removal.

Kinetic type	Kinetic equation	Integrated form	Equation number
Zero-order	$Rc = dc/dt = -k_0$	$C_e - C_0 = -K_0 t$	(9)
First-order	$Rc = dc/dt = -k_1 c$	$\ln (C_e/C_0) = -K_1 t$	(10)
Second-order	$Rc = dc/dt = -k_2 c^2$	$(1/C_e) - (1/C_0) = K_2 t$	(11)

the initial concentration of ATN, pH, PS, time, and intensity of electric current. The impact factor of each parameter is visible in Figure 6.

3.5. Effect of Confounders. In this study, we used humic acid (HA), chlorine ions, and nitrate ions to determine the relative effect of the presence of interfering factors on the removal efficiency of atenolol. The results can be seen in Figure 7.

In the presence of Cl ions in solution, according to equation (8), it acts as a free radical and consumes SO_4^{2-} ions. Therefore, following the decrease in the concentration of SO_4^{2-} ions, the rate of oxidation in the system and finally the removal efficiency of ATN is greatly reduced [34].

$$SO_4^- + Cl^- \longrightarrow SO_4^{2-} + Cl^0 k = 3.1 \times 10^8 \frac{1}{ms}. \quad (8)$$

Also, the presence of nitrate free radicals has a similar effect on the oxidation and final efficiency of the process, but its effect is much less than chlorine [34].

The presence of HA in the ATN removal system by the sono-electro persulfate process led to a relative reduction in the efficiency of this process.

This phenomenon may be due to the tendency of HA to react with free radicals, which reduces the power of SO_4^{2-} ions as the most important free radical affecting the removal process and reduces the process efficiency [35].

3.6. Kinetics of the Model. The removal of ATN from water by a sono-electro-persulfate system has been analyzed by the reaction time function in zero-, first-, and second-order kinetic models, and the results are summarized in Table 3.

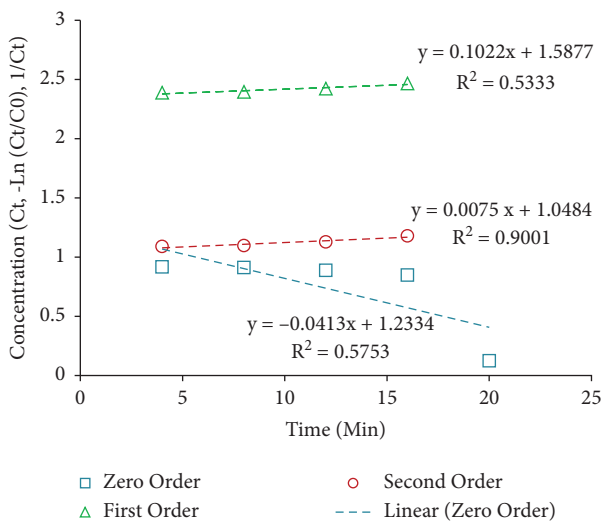


Figure 8: Diagram of nonlinear processing kinetics studied.

Table 4: Constant coefficients of ATN removal reaction kinetics.

Kinetic	Kinetic constant	Amount
Zero-order	K_0	−0.0413
	R^2	0.57
First-order	K_1	0.1022
	R^2	0.53
Second-order	K_2	0.0075
	R^2	0.90

According to Figure 8 and the results shown in Table 4, we found that the second-order model presented is better for the removal of ATN than the other two models.

4. Conclusion

The study contains useful and acceptable information for the practical application of the use of the US/PS/Electricity combination in the removal of ATN. The results show that ATN concentration and contact time are significantly effective in this process and also that neutral pH is the best case for removal of the contaminant by this process. Also, in the optimal conditions, including pH of 7.4, time of 18 min, PS concentration of 2000 mg/L, current intensity of 3.35 A, and an initial ATN concentration of 11.2 mg/L, the removal efficiency of ATN was up to 99.8%.

Based on the results, the method can be considered as a suitable, new, and available way to remove ATN from aqueous solutions.

Data Availability

The data generated and analyzed during this study are available from the corresponding author upon request.

Ethical Approval

The ethics code of this research is IR.MUI.RESEARCH.REC.1398.651.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

Nasrin Zahedi handled investigation and data curation and wrote the original draft. Bahare Dehdashti contributed to the concept and conducting the study and wrote the original draft. Farzaneh Mohammadi was in charge of investigation, modeling, and revising the manuscript. Maryam Razaghi handled the investigation and revised the manuscript. Zeynab Moradmand contributed to conducting the study and revising the manuscript. Mohammad Mehdi Amin handled conceptualization, supervision and methodology and wrote and edited the manuscript.

Acknowledgments

This research was supported and funded by the Student Research Committee of the Isfahan University of Medical Sciences (grant no. 198177).

References

[1] M. Huerta-Fontela, M. T. Galceran, and F. Ventura, "Occurrence and removal of pharmaceuticals and hormones through drinking water treatment," *Water Research*, vol. 45, no. 3, pp. 1432–1442, 2011.

[2] R. Kıdak and Ş. Doğan, "Medium-high frequency ultrasound and ozone based advanced oxidation for amoxicillin removal in water," *Ultrasonics Sonochemistry*, vol. 40, pp. 131–139, 2018.

[3] M. M. Amin, N. Bagheri, F. Mohammadi, and B. Dehdashti, "Sensitivity analysis with the Monte Carlo method and prediction of atenolol removal using modified multiwalled carbon nanotubes based on the response surface method: isotherm and kinetics studies," *International Journal of Chemical Engineering*, vol. 2022, Article ID 4613023, 12 pages, 2022.

[4] M. Yi, Q. Sheng, Q. Sui, and H. Lu, "β-blockers in the environment: distribution, transformation, and ecotoxicity," *Environmental Pollution*, vol. 266, Article ID 115269, 2020.

[5] A. S. Gad El-hak, A. A. Mohammed, A. F. Abdel Hakiem, and R. M. Mahfouz, "Molecular conformation, vibrational spectroscopic and NBO analysis of atenolol and atenolol-hydrochlorothiazide cocrystals," *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, vol. 222, Article ID 117200, 2019.

[6] A. A. Ahmad, A. T. M. Din, N. K. E. Yahaya, J. Karim, and M. A. Ahmad, "Atenolol sequestration using activated carbon derived from gasified Glyricidia sepium," *Arabian Journal of Chemistry*, vol. 13, no. 10, pp. 7544–7557, 2020.

[7] M. M. Amin, B. Dehdashti, L. Rafati, H. R. Pourzamani, M. Mokhtari, and M. Khodadadi, "Removal of atenolol from aqueous solutions by multiwalled carbon nanotubes: isotherm study," *Desalination and Water Treatment*, vol. 133, pp. 212–219, 2018.

[8] S. Hena, L. Gutierrez, and J.-P. Croue, "Removal of pharmaceutical and personal care products (PPCPs) from wastewater using microalgae: a review," *Journal of Hazardous Materials*, vol. 403, Article ID 124041, 2021.

[9] N. Yousefi, S. Pourfadakari, S. Esmaeili, and A. A. Babaei, "Mineralization of high saline petrochemical wastewater using Sonoelectro-activated persulfate: degradation mechanisms and reaction kinetics," *Microchemical Journal*, vol. 147, pp. 1075–1082, 2019.

[10] S. Giannakis, K.-Y. A. Lin, and F. Ghanbari, "A review of the recent advances on the treatment of industrial wastewaters by Sulfate Radical-based Advanced Oxidation Processes (SR-AOPs)," *Chemical Engineering Journal*, vol. 406, Article ID 127083, 2021.

[11] Y.-Q. Gao, N. Gao, J. Chen, J. Zhang, and D. Yin, "Oxidation of β-blocker atenolol by a combination of UV light and chlorine: kinetics, degradation pathways and toxicity assessment," *Separation and Purification Technology*, vol. 231, Article ID 115927, 2020.

[12] M. Moradi, Y. Vasseghian, A. Khataee, M. Kobya, H. Arabzade, and E. N. Dragoi, "Service life and stability of electrodes applied in electrochemical advanced oxidation processes: a comprehensive review," *Journal of Industrial and Engineering Chemistry*, vol. 87, pp. 18–39, 2020.

[13] M. Moradi, Y. Vasseghian, H. Arabzade, and A. Mousavi Khaneghah, "Various wastewaters treatment by sono-electrocoagulation process: a comprehensive review of operational parameters and future outlook," *Chemosphere*, vol. 263, Article ID 128314, 2021.

[14] Y.-J. Liu, Y. L. Huang, S. L. Lo, and C. Y. Hu, "Comparing the effects of types of electrode on the removal of multiple pharmaceuticals from water by electrochemical methods," *Water*, vol. 12, no. 9, p. 2332, 2020.

[15] K. Hasani, A. Peyghami, A. Moharrami, M. Vosoughi, and A. Dargahi, "The efficacy of sono-electro-Fenton process for removal of Cefixime antibiotic from aqueous solutions by response surface methodology (RSM) and evaluation of toxicity of effluent by microorganisms," *Arabian Journal of Chemistry*, vol. 13, no. 7, pp. 6122–6139, 2020.

[16] R. J. Groszmann, G. Garcia-Tsao, J. Bosch et al., "Beta-blockers to prevent gastroesophageal varices in patients with cirrhosis," *New England Journal of Medicine*, vol. 353, no. 21, pp. 2254–2261, 2005.

[17] N. Neha, M. H. Islam, S. Baranton, C. Coutanceau, and B. Pollet, "Assessment of the beneficial combination of electrochemical and ultrasonic activation of compounds originating from biomass," *Ultrasonics Sonochemistry*, vol. 63, Article ID 104934, 2020.

[18] F. Mohammadi, M. R. Samaei, A. Azhdarpoor, H. Teiri, A. Badeenezhad, and S. Rostami, "Modelling and optimizing pyrene removal from the soil by phytoremediation using response surface methodology, artificial neural networks, and genetic algorithm," *Chemosphere*, vol. 237, Article ID 124486, 2019.

[19] S. Hadi, E. Taheri, M. M. Amin, A. Fatehizadeh, and T. M. Aminabhavi, "Synergistic degradation of 4-chlorophenol by persulfate and oxalic acid mixture with heterogeneous Fenton like system for wastewater treatment: adaptive neuro-fuzzy inference systems modeling," *Journal of Environmental Management*, vol. 268, Article ID 110678, 2020.

[20] R. Kumar and N. R. J. Hynes, "Prediction and optimization of surface roughness in thermal drilling using integrated ANFIS and GA approach," *Engineering Science and Technology, an International Journal*, vol. 23, no. 1, pp. 30–41, 2020.

[21] J.-T. Kuo, Y.-Y. Wang, and W.-S. Lung, "A hybrid neural–genetic algorithm for reservoir water quality management," *Water Research*, vol. 40, no. 7, pp. 1367–1376, 2006.

[22] C. Burballa, M. Crespo, D. Redondo-Pachon et al., "Factors associated with the compensation of renal function after nephrectomy of a kidney donor," *Nefrologia*, vol. 38, no. 5, pp. 528–534, 2018.

[23] M. Dolatabadi, M. Mehrabpour, M. Esfandyari, H. Alidadi, and M. Davoudi, "Modeling of simultaneous adsorption of dye and metal ion by sawdust from aqueous solution using of ANN and ANFIS," *Chemometrics and Intelligent Laboratory Systems*, vol. 181, pp. 72–78, 2018.

[24] M. Razzaghi, A. Karimi, Z. Ansari, and H. Aghdasinia, "Phenol removal by HRP/GOx/ZSM-5 from aqueous solution: artificial neural network simulation and genetic algorithms optimization," *Journal of the Taiwan Institute of Chemical Engineers*, vol. 89, pp. 1–14, 2018.

[25] APHA, WEF, and AWWA, *Standard Methods for the Examination of Water and Wastewater*, L. S. Clesceri, A. E. Greenberg, and A. D. Eaton, Eds., American Public Health Association, Washington, DC, USA, 20th edition, 1998.

[26] L. Peng, L. Wang, X. Hu et al., "Ultrasound assisted, thermally activated persulfate oxidation of coal tar DNAPLs," *Journal of Hazardous Materials*, vol. 318, pp. 497–506, 2016.

[27] A. Fatehizadeh, E. Taheri, S. Akbarpour Monjermoyi et al., "Investigating the effectiveness of ultrasonication in removing phenol from synthetics," *Journal of Knowledge and Health in Basic Medical Sciences*, vol. 14, no. 3, pp. 8–15, 2019.

[28] A. R. Rahmani, M. Salari, A. Shabanloo, N. Shabanloo, S. Bajalan, and Y. Vaziri, "Sono-catalytic activation of persulfate by nZVI-reduced graphene oxide for degradation of nonylphenol in aqueous solution: process optimization, synergistic effect and degradation pathway," *Journal of Environmental Chemical Engineering*, vol. 8, no. 5, Article ID 104202, 2020.

[29] R. Hu, L. Zhang, and J. Hu, "Investigation of ozonation kinetics and transformation products of sucralose," *Science of the Total Environment*, vol. 603-604, pp. 8–17, 2017.

[30] A. V. Quaresma, B. A. Sousa, K. T. Silva, S. Q. Silva, A. A. Werle, and R. J. Afonso, "Oxidative treatments for atenolol removal in water: elucidation by mass spectrometry and toxicity evaluation of degradation products," *Rapid Communications in Mass Spectrometry*, vol. 33, no. 3, pp. 303–313, 2019.

[31] A. A. M. Alzayd and F. F. Karam, "Adsorption of atenolol drug from aqueous solution by poly (AAM_MA) hydrogel and used in drug delivery system: study kinetic and thermodynamic," *Research Journal of Pharmacy and Technology*, vol. 12, no. 10, pp. 4678–4682, 2019.

[32] A. Alver, E. Basturk, S. Tulun, and I. Simsek, "Adaptive neuro-fuzzy inference system modeling of 2, 4-dichlorophenol adsorption on wood-based activated carbon," *Environmental Progress & Sustainable Energy*, vol. 39, no. 5, Article ID e13413, 2020.

[33] Z. Jiang, J. Hu, X. Zhang et al., "A generalized predictive model for TiO2–Catalyzed photo-degradation rate constants

of water contaminants through artificial neural network," *Environmental Research*, vol. 187, Article ID 109697, 2020.

[34] M. Y. Badi, A. Esrafili, H. Pasalari et al., "Degradation of dimethyl phthalate using persulfate activated by UV and ferrous ions: optimizing operational parameters mechanism and pathway," *Journal of Environmental Health Science and Engineering*, vol. 17, no. 2, pp. 685–700, 2019.

[35] F. Zhu, Y. Wu, Y. Liang, H. Li, and W. Liang, "Degradation mechanism of norfloxacin in water using persulfate activated by BC@ nZVI/Ni," *Chemical Engineering Journal*, vol. 389, Article ID 124276, 2020.