Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2022.28817

272 (2022) 156–166
October

Prediction and optimization of steroid hormone removal parameters from municipal wastewater by ultrasound probe using artificial neural network and genetic algorithm: a review

Nasrin Mousavi Kia[a], Farzaneh Mohammadi[b,c], Hasti Hashemi Nejhad[a,*]

[a]*Department of Civil Engineering, Isfahan University of Technology, Isfahan, Iran, Tel. +98 9131698439; email: hhasheminejad@iut.ac.ir (H.H. Nejhad), Tel. +98 9133028351; email: n.moosavi@cv.iut.ac.ir (N.M. Kia)*
[b]*Department of Environmental Health Engineering, School of Health, Isfahan University of Medical Sciences, Isfahan, Iran, Tel. +98 9134733362; email: fm_1363@hlth.mui.ac.ir (F. Mohammadi)*
[c]*Environment Research Center, Research Institute for Primordial Prevention of Non-communicable Disease, Isfahan University of Medical Sciences, Isfahan, Iran*

Received 29 December 2021; Accepted 5 August 2022

ABSTRACT

Estrogens are one of the micropollutants in the wastewater which are harmful to the aquatic. Because the biological processes in wastewater treatment plants cannot completely remove micropollutants, these compounds are present in wastewater effluents. Therefore, we need a treatment method to remove the hormones from the wastewater. Ultrasound waves are very effective to eliminate the micropollutants. This study is based on an analysis of publications published since 2000. Here, ultrasound-assisted research on the removal of hormones (estrone (E_1) and 17β-estradiol (E_2)) from wastewater were studied, then data was collected from existing papers and the model was applied to them. Hormone removal from the wastewater by ultrasound-assisted was modeled and optimized using a multilayer artificial neural network coupled with a genetic algorithm. A network was designed in multilayer perceptron. Various training algorithms were evaluated, and the Levenberg–Marquardt (LM) algorithm was selected as the best one. The optimal number of neurons in the hidden layer was 12, according to the maximum correlation coefficient (R), the lowest absolute mean error, the lowest mean bias error, and the minimum mean square error. According to the results of genetic algorithm, the optimum performance conditions were determined, and the results showed that increasing pH and power density, increased the efficiency of hormone removal from the wastewater. Finally, sensitivity analysis was performed by the Spearman method.

Keywords: Hormones removal; Ultrasound; Wastewater; Artificial neural network; Genetic algorithm

1. Introduction

Steroid hormones are one of emerging pollutants in water resources which has destructive effects on aquatic animals [1]. One type of steroid hormone is estrogen. The two most important hormones secreted by all humans and animals are estrone (E_1) and 17β-estradiol (E_2) [2].

Steroid hormones are harmful to human and animal health and interfere with their reproduction; thus, they are a primary environmental concern [3]. Studies in rodents have demonstrated that estrogens are carcinogens in various tissues, including the kidneys, liver, uterus, and mammary glands [4]. Although steroid hormone concentrations are modest (0.2–64 ng/L), their negative impacts on the environment are significant [5]. The rising

* Corresponding author.

concentration of hormones in water and wastewater sources has piqued the interest of many academics. Several research on hormones have been attributed to the removal or reduction of hormones in wastewater. The most essential methods to eliminate these hormones are: activated sludge [6], oxidation ditch [7], aeration lagoon [8], nanofiltration [9], activated carbon adsorption [10], water chlorination and photo-Fenton-like degradation [11]. Studies showed that some of these methods do not have acceptable efficacy to reduce the hormones. According to studies, biological treatment can destroy hormones, but some hormones remain in the effluent. The ability to remove hormones is higher in advanced processes, but they face two problems, restrictions on high costs and the production of dangerous by-products. One of the most effective methods of hormone removal which was considered today is the ultrasound method. One of the essential advantages of this method is the lack of by-products [2].

Another challenge in wastewater treatment is its dynamics. Many changes in discharge, concentration, and composition of incoming wastewater are among these cases. Besides, it is mainly impossible to control these changes. Thus, computer modeling and simulations are required to explain, forecast, and regulate processes with complex relationships. In addition to laboratory research, modeling plays a significant role in maximizing information and saving time and money. A model which can better predict the features and performance of a system is undoubtedly superior. Models such as response surface methodology (RSM) and artificial neural networks (ANN) are used to predict complex pollutant removal processes in the environment.

One of the essential steps to achieve high elimination efficiency of hormones is to study the effective parameters in their elimination and optimize these parameters. According to the researchers, the ANN response was very appropriate to analyze the bacterial processes with complex conditions. Artificial neural networks are modern systems and computational methods for machine learning, knowledge display, and finally, the application of acquired knowledge to maximize the output responses of complex systems [12].

A genetic algorithm (GA) is an innovative and optimized search method inspired by Charles Darwin's theory of natural selection. This algorithm represents the theory of natural selection, where the most suitable people are selected to continue the generation and produce children [13].

Because the ultrasound method is a novel and applicable process to remove micropollutants from wastewater [14], it was investigated in many studies [15,16]. However, using ultrasound to remove hormones appears to evaluate in very few studies. Few studies were performed on removing hormones from wastewater by ultrasound. Indeed, no study was done on modeling and optimizing the effective parameters to eliminate them with this process. Therefore, this study aimed to use artificial intelligence to model and optimize the removal of steroid hormones from wastewater by ultrasound. The results of this review were modeled and the interaction of variables, such as the effect of duration of exposure to ultrasound, pH, frequency, ultrasound power, probe cross-sectional area, and reactor size was evaluated on hormone removal efficiency. To predict the removal efficiency of hormones in different conditions, use an artificial neural network coupled with a genetic algorithm to optimize the process parameters.

2. Materials and methods

2.1. Data gathering

We present an overview of the literature to describe the hormone removal using ultrasound processes. This work is based on a literature review covering the period from 2000 to 2021, and used several scientific web bases: Web of Science, Pub med, Scopus, Google Scholar, Google, Pro-Quest, and Science Direct. Different keywords were applied: hormones, E_1, E_2, ultrasound, micropollutants, and wastewater. Table 1 shows the physico-chemical characteristics of the hormones evaluated in this investigation. As indicated in Table 2, a total of 12 papers evaluated the elimination of micropollutants by ultrasound, three of which dealt with steroid hormones. The following three articles have been mined for information. Fig. 1 depicts the flowchart for this evaluation.

2.2. Selected parameters description

Based on the literature review, the following parameters were selected as the most critical parameters in the ultrasound process, which are pH, power density [Eq. (1)], power intensity [Eq. (2)], frequency, and time.

$$P_d = \frac{P(\text{input})}{V} \tag{1}$$

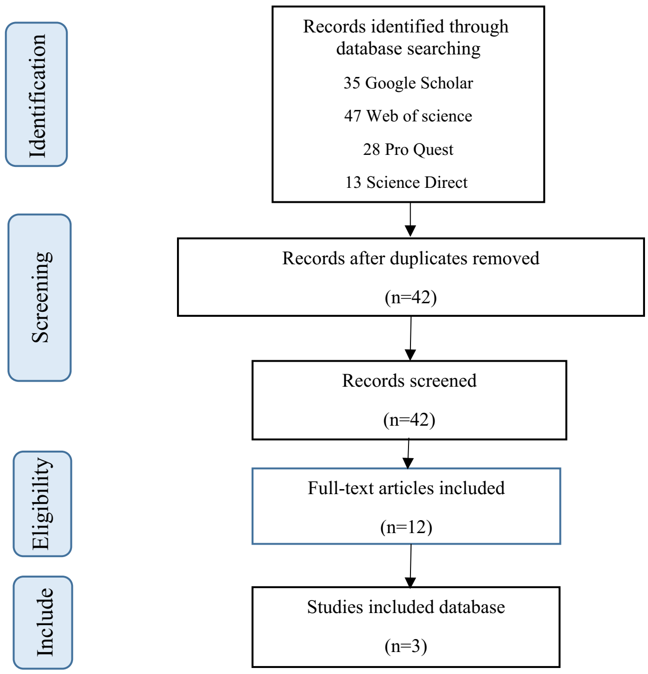


Fig. 1. Review flow diagram of this study.

$$P_i = \frac{P(\text{input})}{A_p} \tag{2}$$

where P_{input}, V, and A_p represent power input, the volume of the sample, and probe transmitting area, respectively.

The independent and responses (dependent) variables and their levels, and descriptive statistics of all input and output parameters are presented in Tables 3 and 4, respectively. Besides, the schematic of ultrasound reactor is given in Fig. 2.

3. Artificial neural networks

3.1. ANN theory

An artificial neural network is a numerical model which imitates the human brain's biological neural network, able to learn and recognize complicated non-linear functions [29,30]. The nntool (a toolbox in MATLAB software), MATLAB version 2014b, have been used for modeling. Feed-forward back-propagation multilayer perceptron ANNs are the most widely used network in environmental engineering [31,32]. There are several layers in a neural network: the input layer, the hidden layer, and the output layer [33]. First, the input vector is multiplied by the cell weight vector; then, the transfer function works on it. The data are then processed in the hidden layer, and the output layer is formed [34]. Each layer is made up of several neurons. The number of hidden layer neurons effectively achieves a reasonable response [Eqs. (3)–(5)]. If too few neurons are included in the network, it might not be possible to fully detect the signal and variance of a complex data set. In contrast, if too many neurons are used, the ANN has too many parameters and may overfit the data [35]. Weights in the neural network are the most critical factor in converting input to output [36].

$$\frac{2(i+o)}{3} < n_h < i(i+o) - 1 \tag{3}$$

$$0.5i - 2 < n_h < 2i + 2 \tag{4}$$

$$\frac{n}{\alpha(i+o)} < n_h < \frac{n}{\alpha(i+o)} 1 < \alpha < 10 \tag{5}$$

where i, o, and n represent number of inputs, number of outputs, and number of hidden layer neurons, respectively [32,37].

The extracted data, which included 229 data sets, were divided into three subsets of 70% training, 15% validation, and 15% test using a random selection method [38]. In many studies, classification has been done in the same way [39].

In the next step, the errors calculated in the validation step are controlled and are expected to decrease during the training. If the calculated error increases during the validation step, the training will stop. Networks with the highest correlation coefficient (R^2) and the lowest error rate: mean square error (MSE), mean absolute error (MAE), and mean bias error (MBE), are the best networks, Eqs. (6)–(9).

$$\text{MSE} = \frac{1}{n}\sum_{i=1}^{n}\left(Y_{\text{pred.}} - Y_{\text{exp.}}\right)^2 \tag{6}$$

$$R^2 = 1 - \frac{\sum_{i=1}^{n}\left(Y_{\text{exp.}} - Y_{\text{pred.}}\right)^2}{\sum_{i=1}^{n}\left(Y_{\text{exp.}} - \overline{Y_{\text{exp.}}}\right)^2} \tag{7}$$

$$\text{MAE} = \frac{1}{n}\sum_{i=1}^{n}\left|\left(\text{Y}_{\text{exp.}} - \text{Y}_{\text{pred.}}\right)\right| \tag{8}$$

$$\text{MBE} = 1/n\sum_{i=1}^{n}\left(\text{Y}_{\text{exp.}} - \text{Y}_{\text{pred.}}\right) \tag{9}$$

where n, Y_{exp}, Y_{pred}, represent the number of dataset values, the experimental value of the experiment, and the predicted value of the experiment by the model, respectively [40–42].

3.2. GA theory

Genetic algorithm (GA) is one of the evolutionary optimization problem-solving techniques [43]. GA produces an initial population at random and then employs mutation, selection, and intersection operators to develop this population. Children that score genetically higher on the fitness function have a greater probability of passing it on to the next generation with each process iteration. Thus, after

Table 1
Physico-chemical properties of steroid hormones [17]

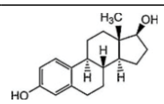
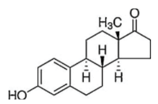
Hormone	Molecular formula	Molecular weight (g/mol)	Water solubility at 25°C (mg/L)	log(K_{oc})	log(K_{ow})	Molecular structure
17β-estradiol (E2)	$C_{18}H_{24}O_2$	272.38	82	2.90	4.01	
Estrone (E1)	$C_{18}H_{22}O_2$	270.37	147	3.02	3.43	

Table 2
Resources studied

No.	Paper name	Parameters	Source	Year	Dataset
1	Review on endocrine-disrupting merging compounds in urban wastewater: occurrence and removal by photocatalysis and ultrasonic irradiation for wastewater reuse	- Concentration (mg/L) - pH - Reaction time - Catalyst and oxidant - Concentrations	[18]	2007	–
2	Ultrasound-assisted destruction of estrogen hormones in aqueous solution: effect of power density, power intensity, and reactor configuration	- pH - Duration of ultrasound exposure - Frequency - Ultrasound power - Probe cross-sectional area - Reactor size	[19]	2007	✓
3	Ultrasound-induced destruction of low levels of estrogen hormones in aqueous solutions	- pH - Duration of ultrasound exposure - Frequency - Ultrasound power - Probe cross-sectional area - Reactor size	[20]	2007	✓
4	Fate of pharmaceuticals in contaminated urban wastewater effluent under ultrasonic irradiation	- pH - Time - Initial concentration of pharmaceuticals - Temperature	[21]	2009	–
5	Processes for the elimination of estrogenic steroid hormones from water: a review	–	[22]	2012	–
6	Determination of phenols and pharmaceuticals in municipal wastewaters from Polish treatment plants by ultrasound-assisted emulsification	- Effect of solvent volume - Impact of manual shaking of sample - Amount of buffering salt - Extraction time	[23]	2013	–
7	Sonochemical techniques to degrade pharmaceutical organic pollutants	- Process mode - Power (W) - Frequency (kHz)	[24]	2015	–
8	Determination of personal care products and hormones in leachate and groundwater from Polish MSW landfills by ultrasound-assisted emulsification microextraction and GC-MS	- Retention time (t_R) [min] - Volume of extraction solvent	[25]	2015	–
9	Degradation of pharmaceuticals by ultrasound-based advanced oxidation process	- Frequency (kHz) - Power - pH - Temp. (°C)	[26]	2016	–
10	Production of hydroxyl free radical, the main the mechanism for removing steroid hormones by ultrasound	- Frequency (kHz) - Power (Watt) - Time (min)	[27]	2016	–
11	Ultrasonic treatment of endocrine-disrupting compounds, pharmaceuticals, and personal care products in water: a review	- pH - Temperature - US frequency - Power - Reactor type - C0 (LG.L_1)	[28]	2017	–
12	Hormones removal from municipal wastewater using ultrasound	- pH - Duration of ultrasound exposure - Frequency - Ultrasound power - Probe cross-sectional area - Reactor size	[2]	2018	✓

several generations, the answer to the problem leads to an optimal solution [44]. GA was used to optimize the parameters included to achieve the highest removal efficiency of E_1 and E_2 hormones.

4. Results and discussion

4.1. Mechanism of the sonochemical process

In the ultrasound process, a series of chemical reactions are caused by sound waves. As a result of these chemical reactions, the cavitation occurs. When ultrasonic waves enter the liquid, they create oscillating zones. In these oscillations, bubbles are formed in terms of positive and negative pressures on the liquid. These bubbles grow due to the absorption of energy by ultrasound waves. Eventually, these bubbles explode during the compression cycle [24]. In a typical treatment, organic pollutants are destroyed in two ways, one by decomposition due to heat generated in the gas and the other by Degrade due to oxidation due to reaction with hydroxyl radicals in gas [45].

Table 3
Amplitude of variables [2,19,20]

Independent variables	Amplitude
pH	3–20
Power density (Pd), W/mL	0.05–2.1
Frequency (f), kHz	20–60
Time (t), min	10–120
Power intensity (Pi), W/cm^2	13–35

4.2. Neural network structure

Using MATLAB neural network, an optimal network with a suitable transmission function was created. This network was trained in different structures. Finally, according to the results obtained in Tables 5 and 6 of the LM (Levenberg–Marquardt) algorithm, showed the maximum correlation coefficient and the minimum error rate. Then a network was created with a hidden layer [46], and then, according to Eqs. (3)–(5), the hidden layer neuron range, with five input parameters and one output parameter, and 229 data, was obtained 12–4, which was trained for each of the numbers in this network range, and finally, 12 neuron showed the lowest error rate. This network was trained using several forms of transfer functions (Tan-Sig, Log-Sig, and Purelin), with the hyperbolic tangent sigmoid function (tansig) used for the hidden layer and a linear function (purelin) used for the output layer, as shown in Table 7. Finally, a neural network with a fixed 5:12:1 topology was developed. Fig. 2 indicates the most effective ANN.

Fig. 3, shows the best correlation coefficient and the lowest error rate for validation, training, and testing data. According to this figure, training the network ends after 11 epochs. Regarding the regression graphs in Fig. 2 show the

Table 4
Descriptive statistics of all input and output parameters based on extracted data

		Hormone removal	pH	Power intensity	Power density	Frequency	Time
Descriptive statistics for E_1 hormone data							
No.	Valid	229	229	229	229	229	229
Mean		47/7	6/69	27/9	0/12	43/6	72/34
Std. error of mean		1/8	0/185	0/4	0/02	0/9	2/287
Median		40/7	7	22/3	0/07	45	60
Mode		15/60[a]	7	22/29[a]	0/05[a]	30[a]	30
Variance		760/4	7/795	49/3	0/092	175/718	1197/2
Minimum		10/1	3	13/48	0/05	20	10
Maximum		98	10	35/03	2/10	60	120
Sum		10,913/93	1,531	6,379/3	28/88	9,980	16,565
Descriptive statistics for E_2 hormone data							
No.	Valid	229	229	229	229	229	229
Mean		46/97	6/69	27/86	0/13	43/58	72/34
Std. error of mean		1/81	0/18	0/46	0/02	0/87	2/29
Median		40/04	7	22/3	0/07	45	60
Mode		11/90[a]	7	22/29[a]	0/05[a]	30[a]	30
Std. deviation		27/45	2/8	7/02	0/3	13/25	34/61
Variance		753/74	7/8	49/27	0/09	175/72	1,197/3
Minimum		10.40	3	13.48	0/05	20	10
Maximum		98.00	10	35.03	2/10	60	120
Sum		10,757/19	1,531	6,379/30	28/88	9,980	16,565

Table 5
Best training algorithm and the optimal number of neurons in the hidden layer for the E_1 hormone

ANN topology	Training algorithm	*R* (all data)	*R* (test data)	MSE	MAE	MBE
5:12:1	Quasi-Newton (BFG)	0.990	0.954	0.00068	0.02125	–0.00115
5:12:1	Conjugate gradient with Powell-Beale restarts (CGB)	0.992	0.990	0.00180	0.02497	–0.00067
5:12:1	Fletcher-Reeves conjugate gradient (CGF)	0.985	0.958	0.00152	0.03010	0.00216
5:12:1	Polak-Ribiere conjugate gradient (CGP)	0.982	0.955	0.00144	0.03216	–0.00074
5:12:1	Levenberg–Marquardt (LM)	0.997	0.995	0.00063	0.01725	0.00007
5:12:1	Scaled conjugate gradient (SCG)	0.992	0.992	0.00099	0.02407	0.00066
5:12:1	Resilient backpropagation (RP)	0.992	0.991	0.00137	0.02755	0.00210
5:12:1	Gradient descent (GD)	0.843	0.837	0.01646	0.11436	0.01457
5:12:1		0.997	0.995	0. 00063	0.01725	0.00007
5:11:1		0.996	0.991	0.000538	0.01784	0.048165
5:10:1		0.994	0.996	0.001121	0.02239	0.042101
5:9:1		0.996	0.993	0.000610	0.01834	0.039879
5:8:1	Levenberg–Marquardt (LM)	0.993	0.979	0.000488	0.02030	0.03384
5:7:1		0.990	0.985	0.001206	0.02418	0.032606
5:6:1		0.995	0.997	0.002283	0.0182	0.026304
5:5:1		0.989	0.967	0.001292	0.02293	0.0152
5:4:1		0.990	0.993	0.000443	0.02293	0.019487

Table 6
Best training algorithm and the optimal number of neurons in the hidden layer for the E_2 hormone

ANN topology	Training algorithm	*R* (all data)	*R* (test data)	MSE	MAE	MBE
5:12:1	Quasi-Newton (BFG)	0.988	0.951	0.00139	0.02134	–0.00225
5:12:1	Conjugate gradient with Powell-Beale restarts (CGB)	0.992	0.993	0.00104	0.02659	–0.00322
5:12:1	Fletcher-Reeves conjugate gradient (CGF)	0.982	0.945	0.00218	0.03061	–0.00178
5:12:1	Polak-Ribiere conjugate gradient (CGP)	0.966	0.921	0.00226	0.05008	–0.00155
5:12:1	Levenberg–Marquardt (LM)	0.997	0.996	0.00045	0.01496	–0.00157
5:12:1	Scaled conjugate gradient (SCG)	0.982	0.983	0.00101	0.02974	–0.0021
5:12:1	Resilient backpropagation (RP)	0.972	0.987	0.00090	0.02887	0.008163
5:12:1	Gradient descent (GD)	0.884	0.869	0.03224	0.096441	–0.01306
5:12:1		0.997	0.996	0. 00045	0.01496	–0.00157
5:11:1		0.996	0.991	0. 000538	0.01784	0.04619
5:10:1		0.994	0.996	0.001121	0.02239	0.04401
5:9:1		0.996	0.993	0.000610	0.01834	0.03622
5:8:1	Levenberg–Marquardt (LM)	0.993	0.979	0.000488	0.02030	0.04027
5:7:1		0.990	0.985	0.001206	0.02418	0.03085
5:6:1		0.995	0.997	0.002283	0.0182	0.02465
5:5:1		0.989	0.967	0.001292	0.02293	0.01251
5:4:1		0.990	0.993	0.000443	0.02293	0.01712

network output for training, validation, and experimental data along a 45° line, it means that the network followed the target well, and the network response was satisfactory.

4.3. Sensitivity analysis

Sensitivity analysis shows how much the output of a network is affected by each of its inputs [47]. SPSS software was used for sensitivity analysis. Sensitivity analysis with Spearman correlation coefficient showed a confidence level of 99%. At this stage, all input parameters in the sensitivity analysis were estimated, and the results were presented in the form of a tornado graph in Fig. 4. Based on these graphs, it was shown that pH, power density, and power intensity had the highest effect on the removal efficiency of E_1 and E_2 hormones. In contrast, the effect of

Table 7
Transfer functions investigation

ANN topology	Transfer function in the hidden layer	Transfer function in the output layer	LM algorithm		
			R (test data)	*R* (all data)	MSE
5:12:1	Tan-sigmoid	Tan-sigmoid	0.996	0.992	0.000774
5:12:1	Tan-sigmoid	Purelin	0.995	0.997	0.00063
5:12:1	Log-sigmoid	Log-sigmoid	0.916	0.921	0.06283
5:12:1	Log-sigmoid	Purelin	0.988	0.994	0.000630
5:12:1	Purelin	Purelin	0.940	0.940	0.006122
5:12:1	Tan-sigmoid	Log-sigmoid	0.931	0.919	0.040323

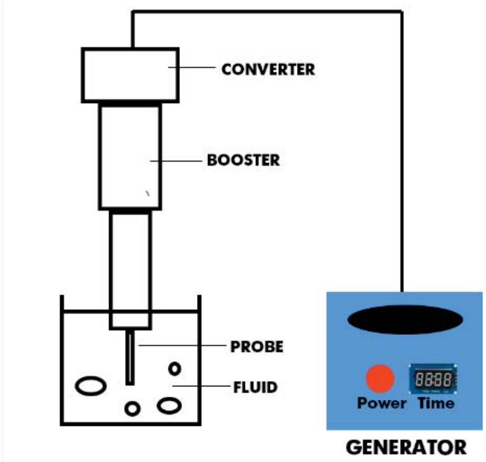


Fig. 2. Schematic of reactor.

time and frequency was negligible, and these results were consistent with neural network results.

4.4. Optimal point by genetic algorithm

Optimization is a procedure of finding and comparing feasible solutions until no better solution can be found [48].

Using GA coding in MATLAB 2014 software, the optimal point of the system for the removal of hormones by ultrasound has been obtained.

Fig. 5 shows the GA response for the highest removal efficiencies of E_1 and E_2 hormones. This result is obtained by considering the neural network trained in the previous sections as a fitness function. The E_1 removal efficiency vs. time at pH 8.95, power intensity 14.75 (W/cm^2), power density 1.26 (W/mL), and frequency 47.79 (kHz) had the highest removal efficiency Fig. 4A). The E_2 removal efficiency against time at pH 9.71, power intensity 33.13 (W/cm^2), power density 0.6 (W/mL), and frequency 42.13 (kHz) had the highest removal efficiency (Fig. 4B).

4.5. Performance evaluation of the ultrasonic process using the ANN-LM model

Because hormones are carcinogenic, their removal from wastewater is crucial for the health of aquatic species and people. The ultrasonic technique is one of the cost-effective, by-product-free processes. In this work, the experimental findings of this approach were modeled using the ANN model, and Figs. 6 and 7 illustrate the ANN-based hormone removal parameters. The plotted levels indicate changes in hormone removal efficiency and are plotted better to understand the effect of the studied removal efficiency parameters. According to Figs. 6A and 7A, hormone removal rate was related to pH nonlinearly, both at low and high reaction times, and the removal efficiency has increased by increasing pH. pH had a more significant effect on removal efficiency than reaction time, and the impact of time was negligible. Figs. 6B and 7B show the effects of power density were almost nonlinear on hormone removal efficiency; power density in values less than 1 W/mL has a great effect on the removal efficiency and increases the removal efficiency, but at values above 1 W/mL had no considerable effect on hormone removal. By Figs. 6C and 7C, both power intensity and time, are nonlinear, such that the removal rate was increased by increasing the amount of power intensity and reaction time, as stated in the article by Suri et al. [19]. Furthermore, Fig. 6D, it is shown that reaction time was more effective against frequency. The frequency was linear at high times and had a non-linear effect at low times, and with increasing frequency, the removal efficiency of E_1 increased. By Fig. 7D, reaction time was more effective against frequency, and with increasing frequency, the removal efficiency of E_2 increased.

Therefore, the most critical parameters to remove hormones in this method are pH and power density. As the initial pH increases, the effect of reducing hormones increases due to the rise in the production of hydroxyl radicals. A similar observation was made by other studies [27].

5. Conclusion

In this study, the effects of pH, frequency, power density, power intensity, and duration on the ultrasonic method's ability to remove steroid hormones from wastewater were examined and modeled using an ANN model. A network with a 5:12:1 structure and LM training algorithm was obtained as the optimal network. This network was used as a fitness function in the genetic algorithm to reach the optimal point. GA results showed that at pH 8.95 and power intensity 14.75 W/cm^2, power density 1.26 W/mL, and frequency 47.79 kHz for hormone E_1 and at pH 9.71, power intensity 33.13 W/cm^2, power density 0.6 W/mL and

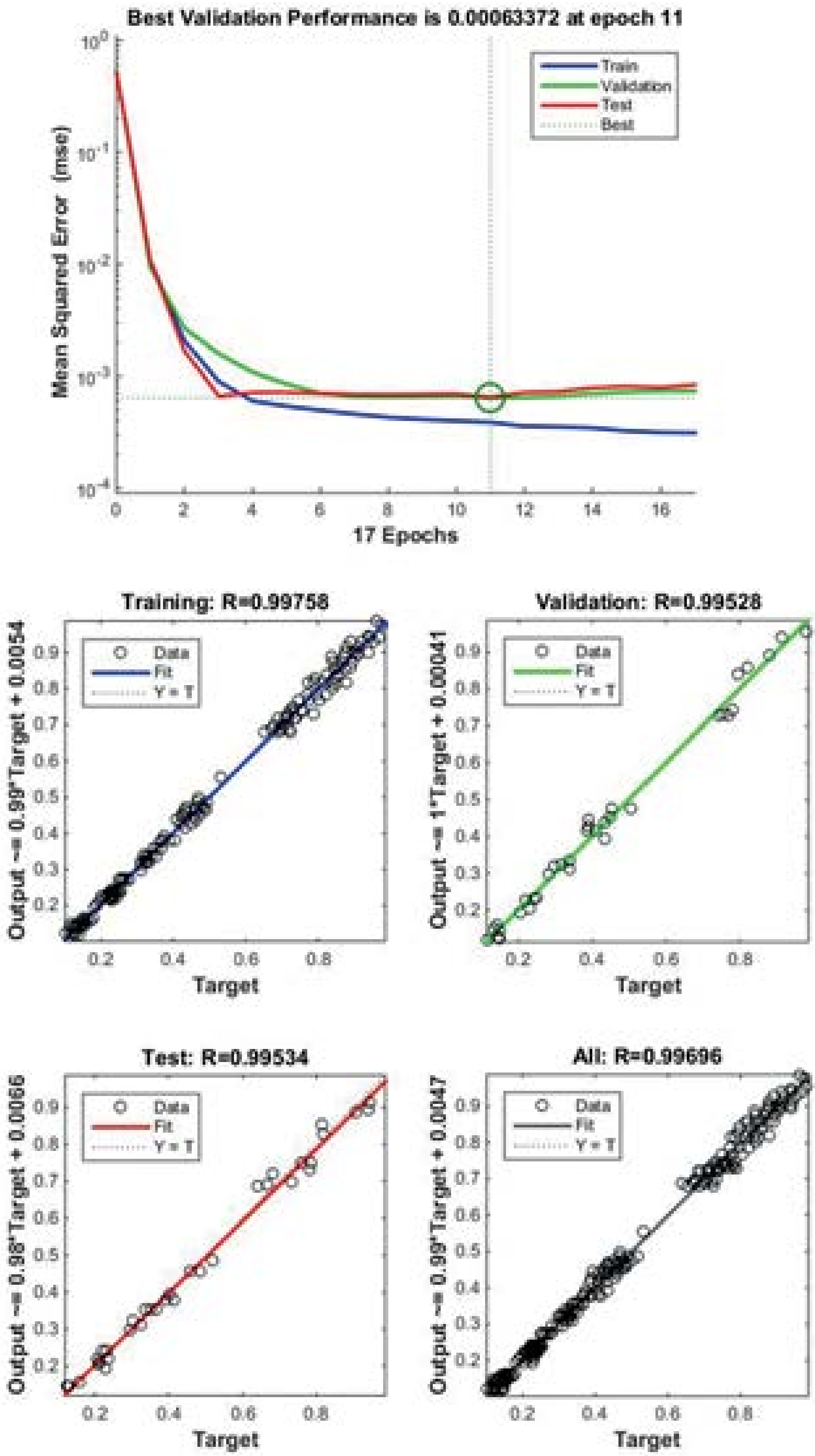


Fig. 3. Regression and MSE error graph of the ANN model.

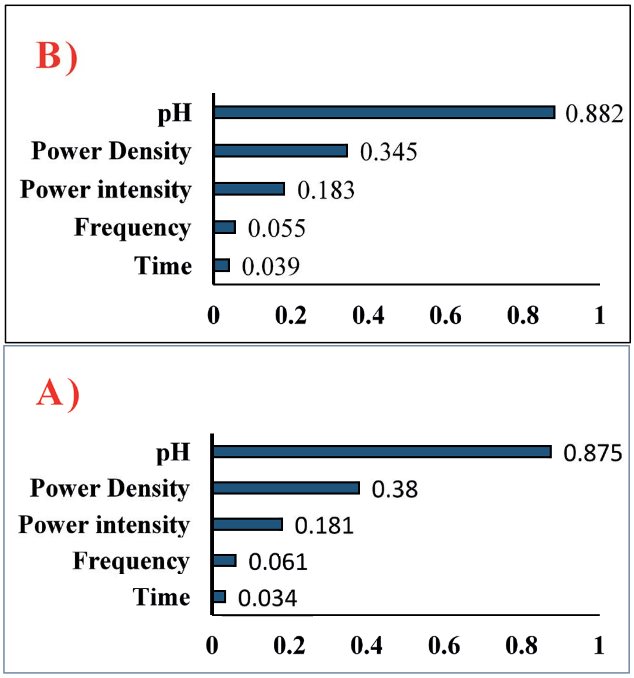


Fig. 4. Tornado graph for sensitivity analysis by standard regression method for (A) E_1 and (B) E_2.

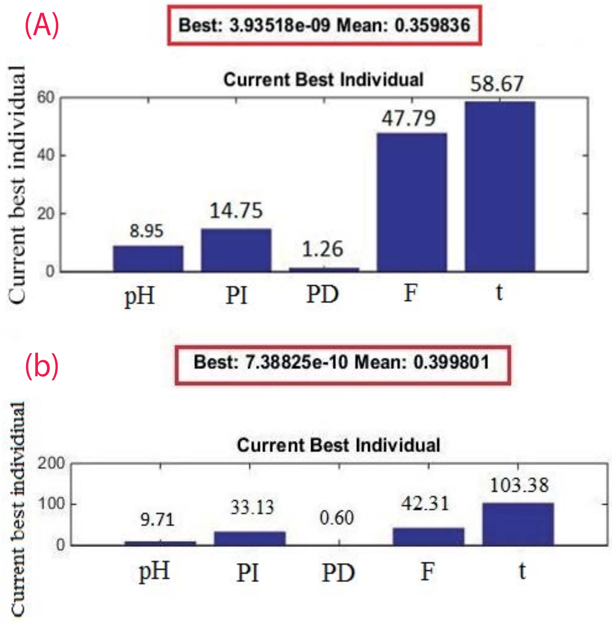


Fig. 5. Genetic algorithm results for: (A) minimum remaining percentage of hormone E_1 and (B) minimum remaining percentage of hormone E_2.

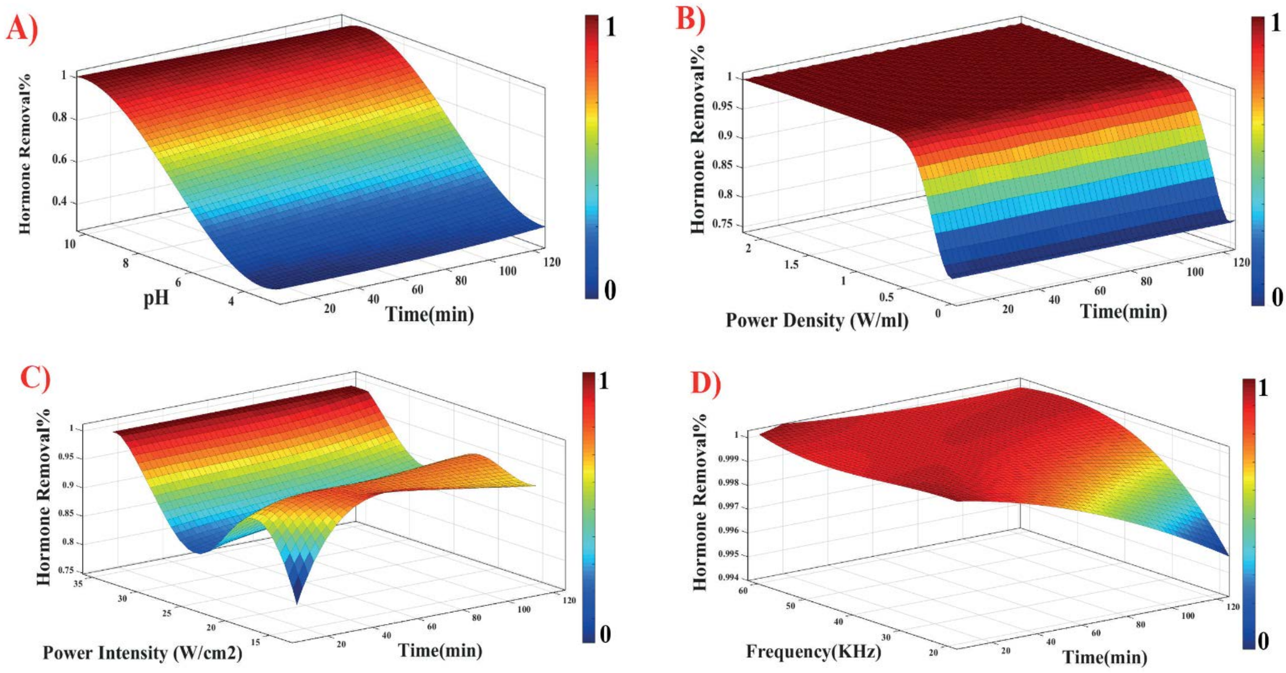


Fig. 6. ANN response surface for (A) pH vs. removal rate of E_1, (B) power density vs. removal rate of E_1, (C) power intensity vs. removal rate of E_1, and (D) frequency vs. removal rate of E_1.

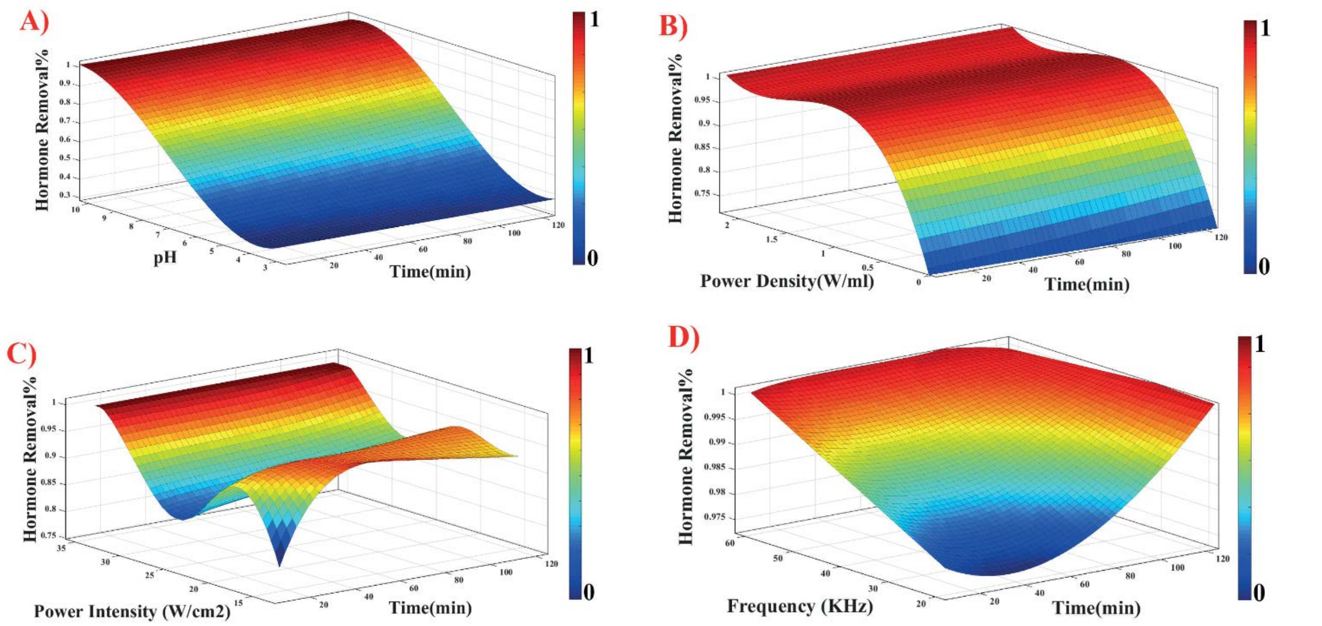


Fig. 7. ANN response surface for (A) pH vs. removal rate of E_2, (B) power density vs. removal rate of E_2, (C) power intensity vs. removal rate of E_2, and (D) frequency vs. removal rate of E_2.

frequency 42.13 kHz for hormone E_2, we reach the highest removal efficiency by ultrasonic method.

Finally, sensitivity analysis was performed with Spearman correlation coefficient and 99% response level and showed that pH is the most influential parameter in hormone removal efficiency.

The results showed that the hormones could be effectively eliminated via ultrasound, significantly when enhancing pH by about 9.

Acknowledgment

Thanks to the Isfahan University of Technology for supporting this thesis; the authors thank Fereidoon Iranpoor for helpful comments in some of the analyses.

References

[1] H. Sirén, S. El Fellah, Steroids contents in waters of wastewater purification plants: determination with partial-filling micellar electrokinetic capillary chromatography and UV detection, Int. J. Environ. Anal. Chem., 96 (2016) 1003–1021.

[2] A. Roudbari, M. Rezakazemi, Hormones removal from municipal wastewater using ultrasound, AMB Express., 8 (2018) 91, doi: 10.1186/s13568-018-0621-4.

[3] N.A. Al-Odaini, M.P. Zakaria, M.I. Yaziz, S. Surif, N. Kannan, Occurrence of synthetic hormones in sewage effluents and Langat River and its tributaries, Malaysia, Int. J. Environ. Anal. Chem., 93 (2013) 1457–1469.

[4] S. Wang, W. Huang, G. Fang, Y. Zhang, H. Qiao, Analysis of steroidal estrogen residues in food and environmental samples, Int. J. Environ. Anal. Chem., 88 (2008) 1–25.

[5] R.G. Maliva, T.M. Missimer, F.P. Winslow, R. Herrmann, Aquifer storage and recovery of treated sewage effluent in the Middle East, Arabian J. Sci. Eng., 36 (2011) 63–74.

[6] B.G. Plósz, H. Leknes, H. Liltved, K.V. Thomas, Diurnal variations in the occurrence and the fate of hormones and antibiotics in activated sludge wastewater treatment in Oslo, Norway, Sci. Total Environ., 408 (2010) 1915–1924.

[7] K. Sornalingam, A. McDonagh, J.L. Zhou, Photodegradation of estrogenic endocrine disrupting steroidal hormones in aqueous systems: progress and future challenges, Sci. Total Environ., 550 (2016) 209–224.

[8] X. Li, W. Zheng, W.R. Kelly, Occurrence and removal of pharmaceutical and hormone contaminants in rural wastewater treatment lagoons, Sci. Total Environ., 445–446 (2013) 22–28.

[9] H. Li, J. Ni, Treatment of wastewater from *Dioscorea zingiberensis* tubers used for producing steroid hormones in a microbial fuel cell, Bioresour. Technol., 102 (2011) 2731–2735.

[10] R. Foroutan, H. Esmaeili, M. Abbasi, M. Rezakazemi, M. Mesbah, Adsorption behavior of Cu(II) and Co(II) using chemically modified marine algae, Environ. Technol., 39 (2018) 2792–2800.

[11] A.O. Ifelebuegu, J. Ukpebor, B. Nzeribe-Nwedo, Mechanistic evaluation and reaction pathway of UV photo-assisted Fenton-like degradation of progesterone in water and wastewater, Int. J. Environ. Sci. Technol., 13 (2016) 2757–2766.

[12] W. Liu, G. Guo, F. Chen, Y. Chen, Meteorological pattern analysis assisted daily PM2.5 grades prediction using SVM optimized by PSO algorithm, Atmos. Pollut. Res., 10 (2019) 1482–1491.

[13] Y. Ding, W. Zhang, L. Yu, K. Lu, The accuracy and efficiency of GA and PSO optimization schemes on estimating reaction kinetic parameters of biomass pyrolysis, Energy, 176 (2019) 582–588.

[14] B. Albero, C. Sánchez-Brunete, A.I. García-Valcárcel, R.A. Pérez, J.L. Tadeo, Ultrasound-assisted extraction of emerging contaminants from environmental samples, TrAC, Trends Anal. Chem., 71 (2015) 110–118.

[15] V. Naddeo, M.F.N. Secondes, L. Borea, S.W. Hasan, F. Ballesteros Jr., V. Belgiornoa, Removal of contaminants of emerging concern from real wastewater by an innovative hybrid membrane process – ultrasound, adsorption, and membrane ultrafiltration (USAMe®), Ultrason. Sonochem., 68 (2020) 105237, doi: 10.1016/j.ultsonch.2020.105237.

[16] T. Benabdallah El-Hadj, J. Dosta, R. Márquez-Serrano, J. Mata-Alvarez, Effect of ultrasound pretreatment in mesophilic and thermophilic anaerobic digestion with emphasis on naphthalene and pyrene removal, Water Res., 41 (2007) 87–94.

[17] H. Ilyas, E.D. van Hullebusch, A review on the occurrence, fate and removal of steroidal hormones during treatment with different types of constructed wetlands, J. Environ. Chem. Eng., 8 (2020) 103793, doi: 10.1016/j.jece.2020.103793.
[18] V. Belgiorno, L. Rizzo, D. Fatta, C.D. Rocca, G. Lofrano, A. Nikolaou, V. Naddeo, S. Meric, Review on endocrine disrupting-emerging compounds in urban wastewater: occurrence and removal by photocatalysis and ultrasonic irradiation for wastewater reuse, Desalination, 215 (2007) 166–176.
[19] R.P.S. Suri, M. Nayak, U. Devaiah, E. Helmig, Ultrasound assisted destruction of estrogen hormones in aqueous solution: effect of power density, power intensity and reactor configuration, J. Hazard. Mater., 146 (2007) 472–478.
[20] H. Fu, R.P.S. Suri, R.F. Chimchirian, E. Helmig, R. Constable, Ultrasound-induced destruction of low levels of estrogen hormones in aqueous solutions, Environ. Sci. Technol., 41 (2007) 5869–5874.
[21] V. Naddeo, S. Meriç, D. Kassinos, V. Belgiorno, M. Guida, Fate of pharmaceuticals in contaminated urban wastewater effluent under ultrasonic irradiation, Water Res., 43 (2009) 4019–4027.
[22] C.P. Silva, M. Otero, V. Esteves, Processes for the elimination of estrogenic steroid hormones from water: a review, Environ. Pollut., 165 (2012) 38–58.
[23] U. Kotowska, J. Kapelewska, J. Sturgulewska, Determination of phenols and pharmaceuticals in municipal wastewaters from Polish treatment plants by ultrasound-assisted emulsification–microextraction followed by GC–MS, Environ. Sci. Pollut. Res., 21 (2014) 660–673.
[24] N. Tran, P. Drogui, S.K. Brar, Sonochemical techniques to degrade pharmaceutical organic pollutants, Environ. Chem. Lett., 13 (2015) 251–268.
[25] J. Kapelewska, U. Kotowska, K. Wiśniewska, Determination of personal care products and hormones in leachate and groundwater from Polish MSW landfills by ultrasound-assisted emulsification microextraction and GC-MS, Environ. Sci. Pollut. Res. Int., 23 (2016) 1642–1652.
[26] M.P. Rayaroth, U.K. Aravind, C.T. Aravindakumar, Degradation of pharmaceuticals by ultrasound-based advanced oxidation process, Environ. Chem. Lett., 14 (2016) 259–290.
[27] A.A. Roudbari, Production of hydroxyl free radical, the main mechanism for removing steroid hormones by ultrasound, Int. J. Health Stud., 2 (2016) 1–5.
[28] K.H. Chu, Y.A.J. Al-Hamadani, C.M. Park, G. Lee, M. Jang, A. Jang, N. Her, A. Son, Y. Yoon, Ultrasonic treatment of endocrine disrupting compounds, pharmaceuticals, and personal care products in water: a review, Chem. Eng. J., 327 (2017) 629–647.
[29] G. Lee, J. Bae, S. Lee, M. Jang, H. Park, Monthly chlorophyll-a prediction using neuro-genetic algorithm for water quality management in Lakes, Desal. Water Treat., 57 (2016) 26783–26791.
[30] K. Anupam, S. Dutta, C. Bhattacharjee, S. Datta, Artificial neural network modelling for removal of chromium(VI) from wastewater using physisorption onto powdered activated carbon, Desal. Water Treat., 57 (2016) 3632–3641.
[31] S. Marina, J. Hazard. Mater., (2018).
[32] J. García-alba, J.F. Bárcena, C. Ugarteburu, A. García, Water Res., (2018).
[33] M. Shirani, A. Akbari, M. Hassani, A. Goli, S. Habibollahi, P. Akbarian, Homogeneous liquid-liquid microextraction via flotation assistance coupled with gas chromatography-mass spectrometry for determination of myclobutanil in cucumber, tomato, grape, and strawberry using genetic algorithm, Int. J. Environ. Anal. Chem., 98 (2018) 271–285.
[34] V.D. Nguyen, H.T.H. Nguyen, V. Vranova, L.T.N. Nguyen, Q.M. Bui, T.T. Khieu, Artificial neural network modeling for Congo red adsorption on microwave-synthesized akaganeite nanoparticles: optimization, kinetics, mechanism, and thermodynamics, Environ. Sci. Pollut. Res. Int., 28 (2021) 9133–9145.
[35] R.M. Adnan, O. Kisi, Transfer learning for neural network model in chlorophyll-a dynamics prediction by Wenchong Tian, Zhenliang Liao, and Xuan Wang, Environ. Sci. Pollut. Res., 27 (2020) 30899–30900.
[36] M.R. Gadekar, M. Mansoor Ahammed, Modelling dye removal by adsorption onto water treatment residuals using combined response surface methodology-artificial neural network approach, J. Environ. Manage., 231 (2019) 241–248.
[37] L. Jin, X. Kuang, H. Huang, Z. Qin, Y. Wang, Study on the overfitting of the artificial neural network forecasting model, Acta Meteorol. Sin., 19 (2004) 216–225.
[38] E. Rahnama, O. Bazrafshan, G. Asadollahfardi, Application of data-driven methods to predict the sodium adsorption rate (SAR) in different climates in Iran, Arabian J. Geosci., 13 (2020) 1160, doi: 10.1007/s12517-020-06146-4.
[39] S. Homayoun Aria, A. Gholamreza, N. Heidarzadeh, Eutrophication modelling of Amirkabir Reservoir (Iran) using an artificial neural network approach, Lakes & Reservoirs: Science, Policy and Management for Sustainable Use, 24 (2019) 48–58.
[40] M. Fan, J. Hu, R. Cao, W. Ruan, X. Wei, A review on experimental design for pollutants removal in water treatment with the aid of artificial intelligence, Chemosphere, 200 (2018) 330–343.
[41] X. Leng, J. Wang, H. Ji, Q. Wang, H. Li, X. Qian, F. Li, M. Yang, Prediction of size-fractionated airborne particle-bound metals using MLR, BP-ANN and SVM analyses, Chemosphere, 180 (2017) 513–522.
[42] M.V. Prabhu, R. Karthikeyan, Comparative studies on modelling and optimization of hydrodynamic parameters on inverse fluidized bed reactor using ANN-GA and RSM, Alexandria Eng. J., 57 (2018) 3019–3032.
[43] M. Rahgoshay, S. Feiznia, M. Arian, S. Ali, A. Hashemi, Modeling daily suspended sediment load using improved support vector machine model and genetic algorithm, Environ. Sci. Pollut. Res., 25 (2018) 35693–35706.
[44] C. Sutherland, A. Marcano, B. Chittoo, Artificial Neural Network-Genetic Algorithm Prediction of Heavy Metal Removal Using a Novel Plant-Based Biosorbent Banana Floret: Kinetic, Equilibrium, Thermodynamics and Desorption Studies, M. Eyvaz, E. Yüksel, Eds., Desalination and Water Treatment, InTechOpen, 2018.
[45] M. Moradnia, M. Noorisepehr, M. Salari, M. Darvishmotevalli, Optimization of 2-chlorophenol removal using ultrasound/persulfate: prediction by RSM method, biodegradability improvement of petrochemical refinery wastewater, Arabian J. Sci. Eng., 47 (2022) 6931–6939.
[46] G. Asadollahfardi, A. Taklify, A. Ghanbari, Application of artificial neural network to predict TDS in Talkheh Rud River, J. Irrig. Drain. Eng., 138 (2012) 363–370.
[47] M. Bahrami, M. Akbari, S.A. Bagherzadeh, A. Karimipour, M. Afrand, M. Goodarzi, Develop 24 dissimilar ANNs by suitable architectures & training algorithms via sensitivity analysis to better statistical presentation: measure MSEs between targets & ANN for Fe–CuO/Eg–Water nanofluid, Physica A, 519 (2018) 159–168.
[48] R. Soleimani, N.A. Shoushtari, B. Mirza, A. Salahi, Experimental investigation, modeling and optimization of membrane separation using artificial neural network and multi-objective optimization using genetic algorithm, Chem. Eng. Res. Des., 91 (2013) 883–903.