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The Use of Artificial Neural Network (ANN) for Modeling of Ammonia Nitrogen Removal from Landfill Leachate by the Ultrasonic Process

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Abstract

Background: *The study examined the implementation of artificial neural network (ANN) for the prediction of Ammonia nitrogen removal from landfill leachate by ultrasonic process.*
Methods: *A three-layer backpropagation neural network was optimized to predict Ammonia nitrogen removal from landfill leachate by ultrasonic process. Considering the smallest mean square error (MSE), The configuration of the backpropagation neural network was three-layer ANN with tangent sigmoid transfer function (Tansig) at hidden layer with 14 neurons, linear transfer function (Purelin) at output layer and Levenberg-Marquardt backpropagation training algorithm (LMA).*
Results: *ANN predicted results were very close to the experimental results with correlation coefficient (R2) of 0.993 and MSE 0.000334. The sensitivity analysis showed that all studied variables (Contact time, ultrasound frequency and power and pH) had strong effect on Ammonia nitrogen removal. In addition, pH was the most influential parameter with relative importance of 44.9%.*
Conclusions: *The results showed that neural network modeling could effectively predict Ammonia nitrogen removal from landfill leachate by ultrasonic process.*

Keywords: Modeling; ANN, Ammonia nitrogen, Leachate, Ultrasound.
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Introduction

Increasingly affluent lifestyles and continuing industrial and commercial growth in many countries worldwide in the past decade have been accompanied by rapid increases in both municipal and industrial solid waste production.[1] The sanitary landfill method for the ultimate disposal of solid waste material continues to be widely accepted and used owing to its economic advantages.[2] The generation of leachate remains an inevitable consequence of the practice of waste disposal in sanitary landfills.[3] Leachate is defined as the aqueous effluent generated as a consequence of rainwater percolation through waste, biochemical processes in waste cells, and the inherent water content of waste itself.[4] Leachates may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist of an important group, as well as ammonia-nitrogen, heavy metals, chlorinated organic, and inorganic salts.[5] When leachate moves downward from landfill into the ground water table as a result of infiltrated precipitation, ground water gets contaminated. Similarly, if the waste is buried below the water table, ground water becomes contaminated after compounds are leached from it.[6] Because ground water and surface water are the sources of our potable water, they should be protected from such pollutants, otherwise the cost of treating drinking water will rise and the biodiversity in surface water bodies will be endangered. Because landfills and leachate production cannot be completely avoided, the only thing to do is to reduce leachate production as much as possible and treat the generated ones to eliminate or reduce the level of contamination in them to discharge content levels before releasing into the environment (receiving water bodies).[7]

During recent years, many new methods, such as physicochemical, biological, and biological combined with physicochemical, have been proposed and tested for the leachate treatment.[8]Ultrasonic, as an advanced oxidation process (AOP), can degrade pollutants not only by producing hydroxyl radicals but also by exerting thermal dissociation (pyrolysis) and shear forces.[9] Ultrasound produces hot spots and strong cavitations in an aqueous solution causing shock waves and reactive free radicals by the violent collapse of the capitation bubbles. These effects should contribute to the physical disruption of microbial structures and inactivation as well as the decomposition of toxic chemicals.[10]

Treating of leachate by AOPs is quite complex, because the process is influenced by several factors. Due to the complexity of the process, it is difficult to model and simulate using conventional mathematical modeling.[11] Artificial neural networks (ANNs) are now used in many areas of science and engineering and are considered a promising tool because of their simplicity toward simulation, prediction, and modeling.[12] The advantages of ANN are that the mathematical description of the phenomena involved in the process is not required; less time is required for model development than the traditional mathematical models and prediction ability, with limited numbers of experiments.[13,14] Application of ANNs to solve environmental engineering problems has been reported in many articles.[15] ANNs were applied in biological and physicochemical waste-water treatment.[16] However, few studies on the applications of ANNs in AOPs have been reported. The present work investigated the implementation of ANNs for the prediction of ammonia-nitrogen removal from landfill leachate by the ultrasonic process. The ANN modeling outputs were compared with the experimental data.

Materials and Methods

Samples of landfill leachate were obtained from a municipal landfill site (>10 years old) located in Shahrood (Semnan, Iran). All leachate samples were collected from leachate lift stations or storage tanks, stored at 3°C, and tested within 2 day of collecting the samples. Characteristics of the leachate samples were COD = 5830 mg/l, BOD_5 = 3940 mg/l, NH_3-N = 730 mg/l, and pH of 8. The ammonia-nitrogen concentrations were analyzed with C_{203} 8 Parameter Test Meter (Hanna electronics Co., Ltd.). The pH was measured by Benchtop pH Meters (Cole-Parmer Co., Ltd.). The pH meter was calibrated before each use with pH 3, 7, and 10 buffer solutions. BOD and COD measurements were determined following standard methods 5210 and 5220, respectively. Reagents and standard chemicals were purchased from Hach Co., except the BOD buffer solution, which was prepared according to Standard Method 5210. BOD check standards were performed with each batch of BOD measurements. The results were considered good when the value of the BOD check standard fell within the range of 198 ± 30.5 mg /l. The average ± standard deviation of the BOD check standards for the entire duration of the project was 169 ± 29 mg /l, which demonstrates good results given the inherent variability in BOD measurements. COD check standards were also performed with each batch of COD measurements. A COD standard solution of 1000 mg /l was diluted to 200 and 500 mg /l to ensure the accuracy of COD measurements. The relative difference for calibration check standards (RD_{cal}) is defined as the absolute difference between the check standard concentration and the known concentration, all divided by the known concentration. The RD_{cal} for COD was <10% for the entire duration of the project.[17]

As shown in Fig. 1, a cylindrical shape Plexiglas reactor with total volume of 1 L was prepared for the laboratory experiments. The solution in the reactor was mixed with a magnetic stirrer, while sufficient aeration was provided by a compressor connected to a porous stone located in the bottom of the reactor. The compressor was used to ensure a completely mixed condition in the reactor. The ultrasonic source was a Model UGMA-5000 ultrasound generator with 30, 45, and 60 kHz transducers having a titanium probe with 20 mm diameter. The power input could be adjusted continuously from 60 to 120 W. A leachate sample of 1000 ml was sonicated in a covered cylindrical glass vessel. Aeration was supplied by a Model SALWAT air compressor. The water level inside the surrounding bath was maintained by continuous circulation of cooling water, and subsequently the temperature was maintained constantly at 30 ± 2 °C. Ferrous sulfate ($FeSO_4 \cdot 7H_2O$), sulphuric acid, and hydrogen peroxide (Merck, 30 wt. %) were of analytical grade.

After the optimization by factorial design, the ultrasonic was applied in the treatment of raw leachate using a batch wise mode. At first, the raw leachate sample was filtered by filter paper (0.45 µ) to remove any suspended solid impurities. Then the sample was adjusted to the required pH with H_2SO_4 or NaOH. Next, different scenarios were tested with regard to power intensities of 70 and 110 W, frequencies of 30, 45, and 60 KHz, reaction times of 30, 60, 90, and 120 min, and pH of 3, 7, and 10. Ammonia-nitrogen concentration of the sonicated sample was measured using Standard Methods 4500. The pH was measured by Model Benchtop pH Meters.

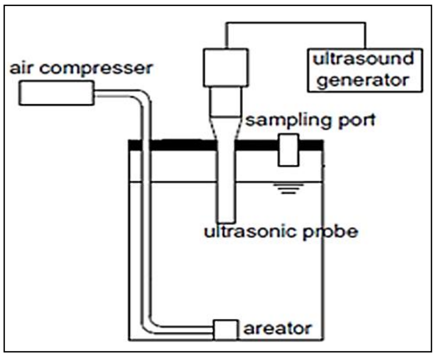

Figure 1. Scheme of the experimental set-up

ANNs are known for their ability of learning, simulation, and prediction of data [18]. The inspiration of using a neural network came from the biology of the human brain. The disadvantage of an ANN is its black box nature [19]. The individual relations between the input variables and the output variables are not developed by engineering judgment, so the model tends to be a black box.[20] Furthermore, there is a greater computational burden and proneness to over fitting and the sample size has to be large.[21] The network consists of numerous individual processing units called neurons commonly interconnected in a variety of structures.[22] The strength of these interconnections is determined by the weight associated with neurons. The multilayer feed-forward net is a parallel interconnected structure consisting of input layer and includes independent variables, number of hidden layers and output layer.[23] Here, a three-layered back propagation (BP) neural network with tangent sigmoid transfer function (Tansig) at hidden layer and a linear transfer function (Purelin) at output layer was used. The BP algorithm was used for network training. Neural Network Toolbox $V_{4.0}$ of MATLAB mathematical software was used for prediction of ammonia-nitrogen removal. Data sets (216 experimental sets) were obtained from our study and were divided into input matrix [p] and target matrix [t]. The input variables were reaction time (t), ultrasound frequency, and power and pH. The corresponding ammonia-nitrogen removal per cent was used as a target. To ensure that all variables in the input data are important, principal component analysis was performed as an effective procedure for the determination of input parameters (Hernandez Ramirez et al., 2014). It was observed that all input variables were important. The data sets were divided into training (one half), validation (one fourth), and test (one fourth) subsets, each of which contained 108, 54, and 54 samples, respectively.

Results

To determine the best BP training algorithm, 10 BP algorithms were studied. Tansig at hidden layer and a linear transfer function (Purelin) at output layer were used. In addition, five neurons were used in the hidden layer as initial value for all BP algorithms. Table 1 shows a comparison of different BP training algorithms. Levenberg-Marquardt BP algorithm (LMA) was able to have smaller mean square error

(MSE) compared to other BP algorithms.[24] Therefore, LMA was considered the training algorithm in the present study.

The optimum number of neurons was determined based on the minimum value of MSE of the training and prediction set.[25] The optimization was done by using LMA as a training algorithm and varying neuron numbers in the range 1-20. Fig. 2 shows the relationship between number of neurons and MSE. MSE was 0.303528 when one neuron was used and decreased to 0.000334 when 14 neurons were used. Increasing neurons to more than 14 did not significantly decrease MSE. Hence, 14 neurons were selected as the best number of neurons. Fig. 3 shows the optimized neural network structure. It has three-layer ANNs, with tansig at hidden layer, with 14 neurons, and linear transfer function (Purelin) at output layer.

The data sets were used to feed the optimized network in order to test and validate the model. Fig. 4 shows a comparison between experimental ammonia-nitrogen removal values and predicted values using the neural network model. The figure contains two lines, one is the perfect fit y=X (predicted data = experimental data), and the other is the best fit indicated by a solid line with best liner equation y=(0.994) X + 0.165, correlation coefficient (R^2) 0.993 and MSE 0.000334. This agrees well with the R^2 reported in the literature–a R^2 of 0.985 for prediction of nitrogen oxides removal by TiO_2 photo catalysis [26], 0.998 for prediction of methyl tert-butyl ether by UV/H_2O_2 process [27], 0.966 for prediction of polyvinyl alcohol degradation in aqueous solution by the photo-Fenton process,[27] 0.995 for removal of humic substances from the aqueous solutions by ozonation,[29] and 0.98 for decoloration of Acid Orange 52 dye by UV/H_2O_2 process.[30]

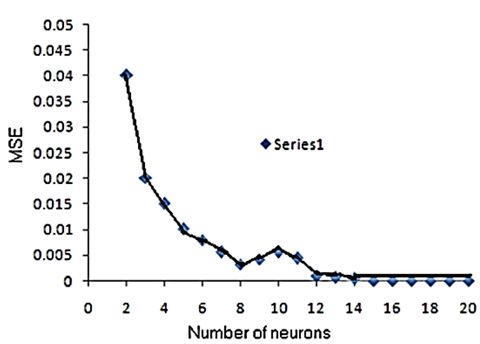

Figure 2. Relationship between number of neurons and MSE

In order to assess the relative importance of the input variables, two evaluation processes were used. The first one was based on the neural net weight matrix and Garson equation.[31] He proposed an equation (Eq. 1) based on the partitioning of connection weights:

$$Ij = \frac{\sum_{m=1}^{m=Nh}\left(\left(\left|W_{jm}^{ij}\right|\right) / \sum_{k=1}^{Ni}\left|W_{km}^{ih}\right|\right)\times\left|W_{mn}^{ho}\right|)}{\sum_{k=1}^{k=Ni}\left\{\sum_{m=1}^{m=Nh}\left(\left|W_{km}^{ih}\right| / \sum_{k=1}^{Ni}\left|W_{km}^{ih}\right|\right)\times\left|W_{mn}^{ho}\right|\right.}$$

Where Ij is the relative importance of the jth input variable on the output variable, Ni and Nh are the number of input and hidden neurons, respectively, and Wis connection weight, the superscripts 'I," "h," and "o" refer to input, hidden, and output layers, respectively, and subscripts "k," "m," and "n" refer to input, hidden, and output neurons, respectively.

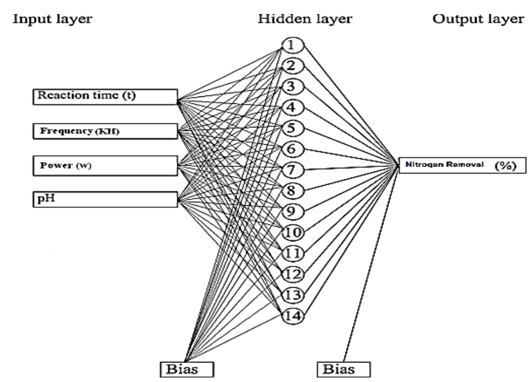

Figure 3. Optimized ANN structure

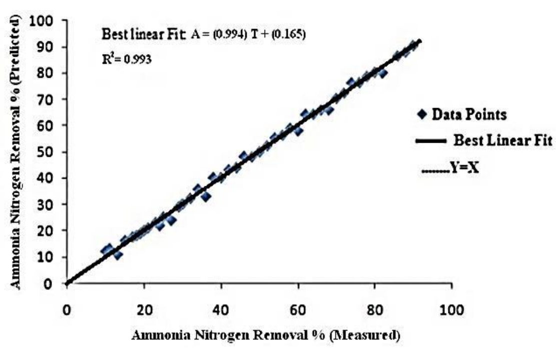

Figure 4. Comparison between predicted and experimental values of the output

Table 2 shows the weights between the artificial neurons produced by the ANN model used in this work. Table 3 shows the relative importance of the input variables calculated by Eq. [1]. All variables have a strong effect on ammonia-nitrogen removal. The pH appears to be the most influential variable followed by power, frequency, and time. The second evaluation process is based on the possible combination of variables. Performances of the groups of one, two, three, and four variables were examined by the optimal ANN structure using the LMA with 14 hidden neurons. The input variables were p_1 (Contact time), p_2 (Frequency), p_3 (Power), and p_4 (pH).

Table 4 shows the results of the sensitivity analysis for different combinations of variables. The sensitivity analysis showed that p_3 was the most effective parameter among other variables in the group of one variable. The MSE (270.21) decreased up 0.304114, which is the minimum value of the group of two variables when p_3 was used in combination with p_2. The MSE (0.304114) decreased up to 0.116543, which is the minimum value of the group of three variables when p_2 was used in combination with p_3 and p_4. The best group performances according to number of parameters are highlighted in Table 4. MSE values decreased as the number of variables in the group increased due to the contribution of all parameters (Table 4). It can be concluded that pH is the most effective parameter. In addition, all variables have a strong effect on ammonia-nitrogen removal and it {2.2 [EN] Subject unclear} agrees well with the sensitivity analysis using the Garson equation.

The pH value influences the generation of hot spots and hydroxyl radicals and hence removal efficiency.[32] To examine the effect of pH, experiments were conducted by varying the pH in the range 3-10. An initial ammonia-nitrogen concentration was 730 mg/l. The other operating conditions were fixed at pH 7, p_3 110 W, and p_2 45 KH. The results (Figure 5) show that pH significantly influences ammonia-nitrogen removal. Decrease in ammonia-nitrogen removal at pH lower than 3 may be due to the decrease in ammonia stripping rate.[32] Furthermore, amount of hydroxyl radicals would decrease at low pH, decreasing degradation of ammonia-nitrogen intermediates. In terms of the relationship between the experimental results and the predicted values of ammonia-nitrogen removal by the model, Figs. 5A-C show that predicted values are in good agreement with the experimental results.

To examine the effect of contact time on ammonia-nitrogen removal, initial contact was varied in the range of 30-120 min at constant initial NH_3 730 mg/L. The corresponding ammonia-nitrogen removal percentage was 64, 72, 78, and 86. The other operating conditions were fixed at pH 7, p_3 110 W, and p_2 45 KH. Figure 6 shows a comparison between the predicted and experimental values of ammonia-nitrogen removal at different contact times. The results show that contact time increases ammonia-nitrogen removal so far due to the increase of cavitations of ammonia nitrogen.[33]

The power value influences the energy of hot spots and cavitations and hence the removal efficiency [34]. To examine the effect of power, experiments were conducted by varying the power in a range of 70 and 110 W. An initial ammonia-nitrogen concentration was 730 mg/l. The other operating conditions were fixed at pH 7 and frequency 45 KH. Figs. 7a-b show a comparison between the predicted and experimental values of ammonia-nitrogen removal at different powers.

The frequency value influences the number of hot spots and cavitation's energy and hence the removal efficiency.[35] To examine the effect of frequency, experiments were conducted by varying the frequency in the range of 30-60 KH. An initial ammonia-nitrogen concentration was 730 mg/l. The other operating conditions were fixed at pH 7 and power 110 W. Figs. 8a-c show a comparison between the predicted and experimental values of ammonia nitrogen removal at different frequencies.

Table 1. Comparison of 10 backpropagation algorithms with 5 neurons in the hidden layer

Backpropagation (BP) algorithm	Function	Mean square error (MSE)	Epoch	Correlation coefficient (R^2)	Best linear equation
Levenberg-Marquardt backpropagation	trainlm	0.00822154	32	0.992	y = 0.992X + 0.325
Scaled conjugate gradient backpropagation	trainscg	0.01675187	95	0.984	y = 0.983X + 0.938
BFGS quasi-Newton backpropagation	trainbfg	0.018652	54	0.988	y = 0.974X + 0.862
One step secant backpropagation	trainoss	0.0306147	29	0.974	y = 0.953X + 1.86
Batch gradient descent	traingd	0.486587	96	0.778	y = 0.365X+16
Variable learning rate back propagation	traingdx	0.449258	65	0.771	y = 0.447X+17
Batch gradient descent with momentum	traingdm	0.508212	104	0.719	y = 0.341X + 19.8
Fletcher–Reeves conjugate gradient backpropagation	traincgf	0.0272147	219	0.968	y = 1.58X−0.369
Polak–Ribi'ere conjugate gradient backpropagation	traincgp	0.0174289	98	0.974	y = 0.974X + 2.34
Powell–Beale conjugate gradient backpropagation	traincgb	0.0203258	39	0.974	y = 0.974X + 0.256

Table 2. Weight matrixes, weights between input and hidden layers (W1) and weights between hidden and output layers (W2)

Neuron	W_1				W_2
	Input variables				Output (Ammonia nitrogen removal %)
	Time	Frequency	Power	pH	
1	0.4789	0.0964	-0.4536	0.4635	0.7479
2	--1.0563	-2.351	0.4261	-1.2454	1.9365
3	0.0598	-0.1704	0.1025	0.4352	-1.5625
4	0.0685	0.0987	-0.0365	0.4795	0.979
5	0.1897	0.6598	0.2653	0.4125	-0.8978
6	-0.2365	-1.4478	0.6658	0.8562	-0.5321
7	0.7896	-0.9863	0.3254	0.6589	-0.8263
8	-0.5362	-0.3125	0.5698	0.8562	-1.1265
9	0.4321	0.1245	0.2365	0.6325	-1.5896
10	-0.352	0.5236	0.5269	0.9987	-0.4563
11	-0.9254	-0.4365	-1.4289	0.8741	-1.1236
12	0.2145	-0.0986	0.1421	0.4653	-1.065
13	0.0236	0.4532	-0.2563	0.4598	-0.3496
14	0.0365	0.8965	1.1123	1.9635	1.7465

Table 3. The relative importance of the input variables

Input variable	Importance %
Time	13
Frequency	19.5
Power	22.6
pH	44.9

Table 4. Evaluation of possible combinations of input variables

Combination	Mean square error (MSE)	Epoch	Correlation coefficient (R^2)	Best linear equation
P1	363.211	8	0.316	Y= 3.69X + 456
P2	270.21	9	0.379	Y= 6.23X + 516
P3	265.341	13	0.567	y = 7.28X + 418
P4	0.563251	7	0.636	y = 6.26X + 614
P1 + P2	0.589421	15	0.413	y = 1.99X+589
P1 + P3	.0521111	10	0.465	y = 0.875X+16.6
P1 + P4	0.390452	8	0.423	y = 0.589X + 16.2
P2 + P3	0.304114	11	0.526	y = 0.656X−21.2
P2 + P4	0.456321	6	0.644	y = 0.656X + 26.8
P3 + P4	0.489652	6	0.689	y = 0.789X + 14.7
P1 + P2 + P3	0.189652	8	0.0.712	y = 0.666X + 15.6
P1 + P2 + P4	0.175421	9	0.897	y = 0.552X + 22.1
P2 + P3 + P4	0.116543	11	0.778	y = 0.778X + 10.3
P1 + P2 + P3 +P4	0.132565	7	0.713	y = 0.663X + 20.5

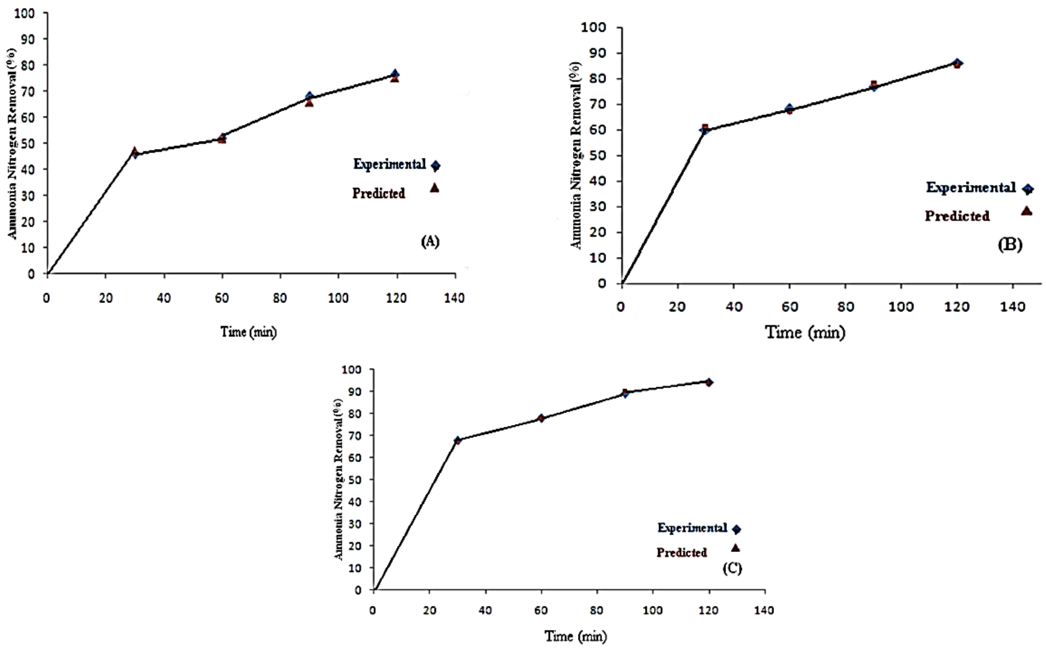

Figure 5. Comparison between ANN output and experimental results at different pH: (A) 3, (B) 7, and (C) 10.

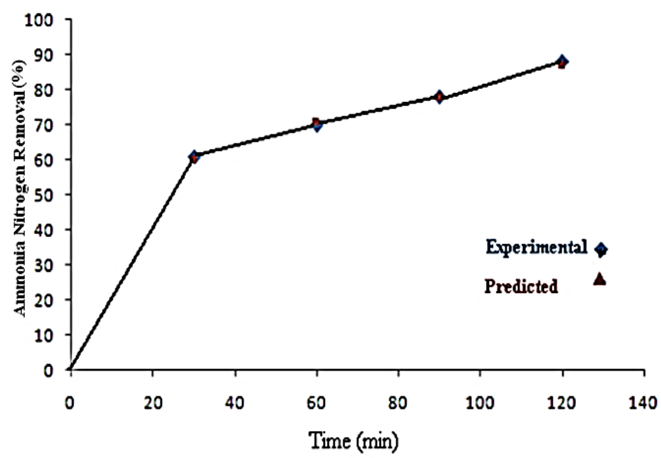

Figure 6. Comparison between ANN output and experimental results at different times

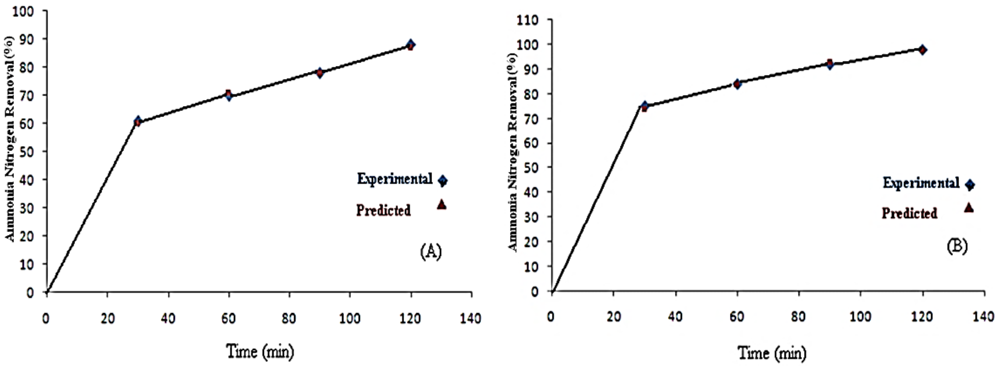

Figure 7. Comparison between ANN output and experimental results at different Powers: 70 (A), 110 (B)

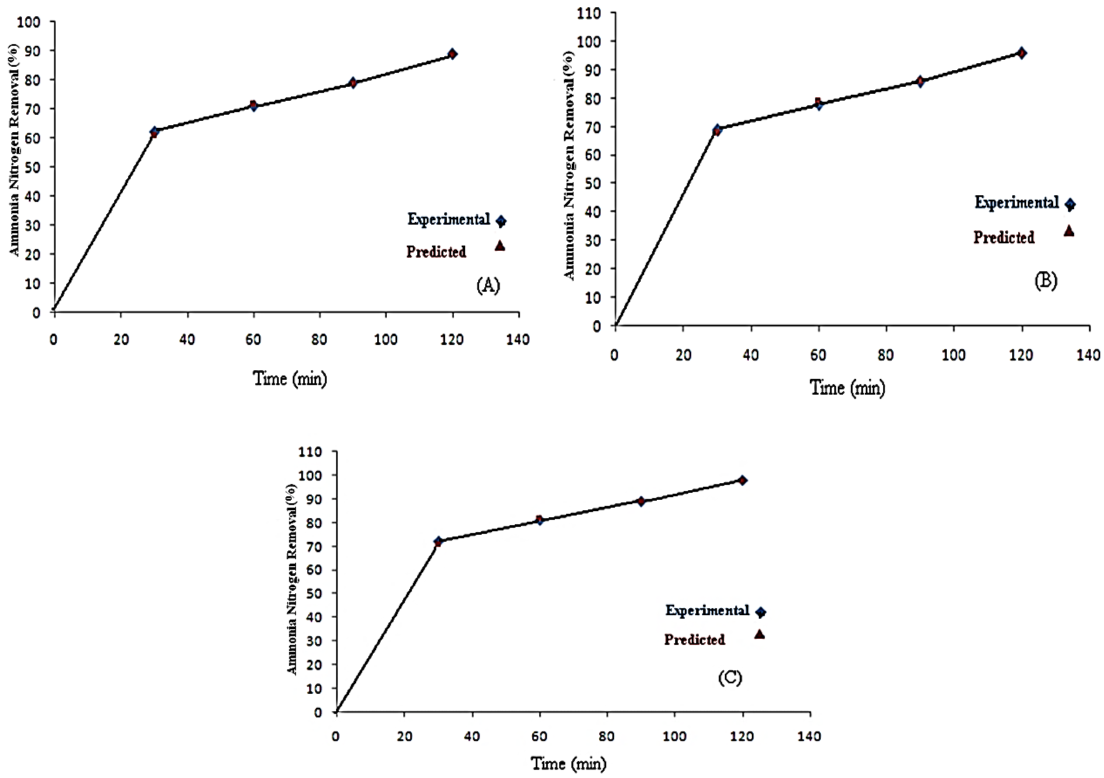

Figure 8. Comparison between ANN output and experimental results at different Frequencies: 30 (A), 45 (B) and 60 (C)

Discussion

A three-layer back propagation neural network was optimized to predict the ammonia nitrogen removal from landfill leachate by ultrasonic process. The configuration of the BP neural network giving the smallest MSE was three-layer ANN with tansig at hidden layer with 14 neurons, linear transfer function (Purelin) at output layer, and LMA. ANN predicted results are very close to the experimental results, with R^2 of 0.993 and MSE 0.000334. The sensitivity analysis showed that all studied variables (contact time, power, frequency, and pH) have a strong effect on ammonia nitrogen removal. In addition, pH is the most influential parameter with a relative importance of 44.9%. ANN results showed that neural network modeling could effectively predict the behavior of the process.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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