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Modeling of simultaneous adsorption of dye and metal ion by sawdust from aqueous solution using of ANN and ANFIS

Maryam Dolatabadi [a], Marjan Mehrabpour [b], Morteza Esfandyari [c,*], Hosein Alidadi [d], Mojtaba Davoudi [e]

[a] Environmental Sciences and Technology Research Center, Department of Environmental Health Engineering, Shahid Sadoughi University of Medical Sciences, Yazd, Iran
[b] Department of Environmental Health Engineering, School of Health, Mashhad University of Medical Sciences, Mashhad, Iran
[c] Department of Chemical Engineering, Faculty of Engineering, University of Bojnord, Bojnord, Iran
[d] Department of Environmental Health Engineering, Mashhad University of Medical Sciences, Mashhad, Iran
[e] Department of Environmental Health Engineering, Health Sciences Research Center, Torbat Heydariyeh University of Medical Sciences, Torbat Heydariyeh, Iran

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ABSTRACT

The current work deals with the investigation of Simultaneous of Basic Red46 (BR46) and Cu (dye and heavy metal) removal efficiency from aqueous solution through the adsorption process using a laboratory scale reactor. In this research, a feed-forward artificial neural network (ANN) and an adaptive neuro-fuzzy inference system (ANFIS) have been utilized to the prediction of adsorption potential of sawdust in simultaneous removal of a cationic dye and heavy metal ion from aqueous solution. Five Operational variables, concluding initial dye, initial Cu (II), pH, contact time, and adsorbent dosage were selected to investigate their effects on the adsorption study. The application of (ANN) and (ANFIS) models for experiments were employed to optimize, create and develop prediction models for dye and Cu (II) adsorption by using sawdust from Melia Azedarach wood. The result reveals that ANN and ANFIS models as a promising predicting technique would be effectively used for simulation of dye and metal ion adsorption. According to this result, in training dataset determination coefficient were obtained 0.99 and 0.98 for dye and a metal ion, respectively. Also, in ANFIS model R^2 was calculated 0.99 for both of pollutants.

1. Introduction

It is known that the Dyes and heavy metals are common and dangerous pollutants due to its toxicity in the aquatic system. Thus, their discharge in large quantities from The textile effluents and wastewater of various industrial processes contains pulp and paper processing, leather tanning, battery production, and other industries to the environment as effluents is one of the most important serious problems that human facing with them(it) [1]. In the industrial effluent, Basic Red 46, introduced in different classifications as a basic, azo, and reactive dye, and copper II is a most common heavy metal ion [2–5]. These pollutants were chosen as a typical dye and heavy metal ions to investigation their simultaneous adsorption in an aqueous solution that has a wide application in textile dyeing processes. There are several methods to remove this pollutant from the aqueous solution. A chemical process such as precipitation, neutralization, electrochemical reduction, ion exchange, cementation, coagulation and flocculation, biosorption, and membrane processes [6–8]. However, there are some disadvantages and limitations. Among them, adsorption is an attractive alternative process due to high efficiency, more economical, easy handling, the available and abundant adsorbents, and also its cost-effectiveness and easiness [9]. The term adsorption refers to a process wherein a material is concentrated at a solid surface from its liquid or gaseous surroundings. Activated carbon is the oldest adsorbent known and is usually prepared from coal, coconut shells, lignite, wood etc., using one of the two basic activation methods: physical and chemical. Activated carbon is among the most effective adsorbents because it has a high surface area which makes greater sorption capacity. However, its wide application is limited because of the high price and hard regeneration. Therefore, it is necessary to seek the cheaper and more available materials as an alternative for activated carbon [10,11]. Numerous studies on low-cost material for dye and heavy metal removal have been reported in recently. A lot of

* Corresponding author. 4th km to road to Esfarayen, University Of Bojnord, Bojnord, Iran
E-mail addresses: health.dolatabadi@gmail.com (M. Dolatabadi), marjanmehrabpour@gmail.com (M. Mehrabpour), M.esfandyari@ub.ac.ir (M. Esfandyari), alidadih@mums.ac.ir (H. Alidadi), davoudi85@gmail.com (M. Davoudi).

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nonconventional, low cost and easily obtainable adsorbents have been tested on a large scale for dye and Cu removal such as sawdust, grap stalk waste, hazelnut, and almond shells, wheat bran, fly ash, tea waste, the carbon of nut shells and rice husk. In recent years, there has been growing interest in finding inexpensive and effective synthetic and natural adsorbents, or their modified products such as montmorillonite, zeolite, chitosan, sawdust and etc. Sawdust is a product of cutting, grinding, drilling, sanding, or otherwise pulverizing wood with a saw or other tool. Sawdust is a readily available wood waste from the carpentry, paper, and furniture industry [12].

Amongst them, Sawdust of Melia Azedarach is a known wood commonly found in subhumid tropical regions of Mashad, Iran where this plant is highly cultivated. It is a lignocellulosic material contains three main components including hemicelluloses, cellulose, and lignin. When sawdust is in contact with water Because of its structure, it takes negative charges on the surface. To remove contaminants from aqueous solution, Sawdust is a relatively abundant, readily available, and inexpensive material commonly being investigated as an adsorbent [13–15].

ANN is an advanced mathematical tool based on the neural structure of the brain. It is composed of many neurons (nodes) that co-operate to perform the desired function. The commonest structures of an artificial neural network consist of three different layers of units: a layer of "inputs" units is connected to a layer of "hidden" units, which is connected to a layer of "outputs" units. Therefore, a biological neuron receives inputs from external sources, combines them in some ways, which represent an intelligently nonlinear activation functions on the result, and then predicts output parameters based on the experimental data in the final result. Recently, ANN has been successfully applied to predict the adsorption behavior in aqueous solution. It is a necessity to have some experimental data for training, validation, and test sets. Many researchers have been reported the different studies in using of artificial neural networks (ANN) approach for modeling of adsorption process in dye [16,17] and Cu adsorption [9,18] separately. Although there is already a considerable amount of research in the adsorption of dye and heavy metal, separately, few studies from different scientific areas have been reported in the field of the application of ANN technique for simultaneous removal of dye and heavy metal.

A branch of artificial intelligence (AI) is Adaptive neuro-fuzzy inference systems (ANFIS) that it is consisting of the learning abilities of artificial neural networks (ANN) and reasoning abilities of fuzzy systems [19–21]. Therefore, it has the advantages of both models in a single methodology. In comparison with using a single methodology by a combination of the fuzzy systems and artificial neural network, an efficient approach is created in many engineering applications. ANFIS is a powerful instrument for modeling, mapping, forecasting, problem-solving, and data mining the input and output values relationship in order to describe nonlinear behavior in complex systems. The structure of an ANFIS model is composed of two parts, namely, containing the antecedent and the conclusion part that are connecting to each other by fuzzy rules in the network form [22–24]. It is widely accepted as a technology due to its universal ability to simulate nonlinear variation, application in the prediction of the performance of many processes, and extrapolation based on historical data in a variety of field [25,26]. Although the previous study by ANFIS model was performed [6,27,28] but a method for removal of simultaneous these pollutants with sawdust by using ANN and ANFIS techniques does not take place in literature.

Therefore, the main motivation behind this investigation is to predict the modeling of the adsorption behavior of sawdust by using both of models (ANN and ANFIS techniques). First, sawdust of Melia Azedarach (waste material) is used as an adsorbent for simultaneous adsorption of dye and Cu (II) from aqueous solution. Then, the operating variables included the initial concentration of dye and Cu (II), solution pH, contact time, and adsorbent dosage are considered as the input data. Finally, output data (percent of dye and Cu (II) removal) calculated from the two models is compared with experimental data in order to further investigate.

2. Materials and methods

2.1. Chemicals

The commercial azo dye Basic Red46 (BR46, cationic dye, ≥98% purity), used without further purification was obtained from a local factory (Shadiloon textile firm, Iran) and used for the preparation of the stock solution. The molecular weight of the dye is 356.84 $gmol^{-1}$, and its maximum absorbance wavelength is at 530 nm. The stock solution of the dye and metal ion used in this study (1000 $mg\,L^{-1}$) were prepared separately by dissolving weighed quantities of BR46 and $CuSo_4.5H_2O$ salts in doubled distilled water (DDW). Other chemical reagents used in the experiment were analytical grade products such as $CuSO_4.5H_2O$ (≥99% purity), HCl (≥37% purity), NaOH (≥99% purity), and KNO_3 (≥99.5% purity) purchased from Merck Company (Darmstadt, Germany). FTIR spectroscopy analysis of the samples was performed to specify the functional groups on the adsorbent in the range of 400–4000 cm^{-1} with FTIR spectrometer (Thermo Nicolet, Avatar 370 model FTIR). A digital pH meter (HACH HQ440D Benchtop Multimeter, LOVELAND, USA) with the glass electrode was used for pH measurements.

2.2. Preparation and modification of adsorbent

The M.azaderach wood, collected from a local garden, was used as an adsorbent. Deadwood of Pollard Melia Azedarach trees is available in all seasons. It was milled and ground to pass through 50 mesh sieves(300μm). To activate the surface sites of pristine sawdust Hydrochloric acid was used. Several studies showed that HCl is a more effective treatment compared with other strong acids such as H_2SO_4 and HNO_3, bases, and alcohols. Five grams of Melia azedarach sawdust (MAS) was soaked with 100 mL of 10% HCl solution. After shaking at 80 °C for 30 min, it was washed with double distilled water to remove the residual acid and then dried at 80 °C for 24 h. Finally, the dried material was ground and sieved to get 50 mesh size particles.[26] To prevent contamination of the final product, it was preserved in an air-tight container for using at subsequent sorption experiments [13].

2.3. Characterization of adsorbent

FTIR spectroscopy analysis of the samples was performed to specify the functional groups on the adsorbent in the range of 400–4000 cm^{-1} with FTIR spectrometer (Thermo Nicolet, Avatar 370 model FTIR). A digital pH meter (HACH HQ440D Benchtop Multimeter, LOVELAND, USA) with the glass electrode was used for pH measurements. The point of zero charges (pHpzc) of the prepared adsorbent was determined in 0.01 M KNO_3 solutions adjusted at different initial pH levels at ambient temperature. The final pH of solutions was measured after 48 h shaking when the equilibrium was reached. In the end, on the plot of final pH values against initial pH values were recorded (pHinitial = pHfinal). The adsorption ability of the surface and the type of surface active centers are indicated by the significant factor that is the point of zero charges (pHpzc). The pH at which the surface charge is zero is called the point of zero charges (pzc) and is typically used to quantify or define the electrokinetic properties of a surface. The value of pH is used to describe pzc only for systems in which H^+/OH^- are the potential determining ions.

In order to determine pH_{pzc}, primarily a 0.01 M KNO_3 solution was prepared. Diluted solutions of HCl or NaOH were applied to adjust the pH of the solution between 3 and 10. The adsorbent (0.3 g) was added to 100 mL of the pH-adjusted solution in an Erlenmeyer flask and agitated with a magnetic shaker for 48 h at room temperature. The final pH of the solution was recorded and plotted against the initial pH. The pH at which the curve crosses the pH initial = pH final line was taken as the pzc.

Table 1
The best R^2 value for BPNN with a different structure.

NO	hidden layer	Dye removal				Cu removal			
		R^2				R^2			
		train	validation	test	total	train	validation	test	total
1	[5]	1.0	0.95	0.96	0.96	0.99	0.99	0.99	0.99
2	[5 5]	0.96	0.84	0.71	0.90	0.87	0.90	0.90	0.87
3	[10]	0.95	0.65	0.79	0.84	1.0	0.95	0.88	0.95
4	[10 10]	0.99	0.93	0.95	0.95	0.98	0.80	0.62	0.84
5	[5 10 5]	0.91	0.98	0.98	0.90	0.9	0.72	0.93	0.90
6	[5 10 10 5]	1.0	0.98	0.95	0.95	0.99	0.93	0.94	0.96
7	[5 10 10 10 5]	0.99	0.96	0.83	0.95	0.99	0.97	0.90	0.94

2.4. ANN and ANFIS method

In this work, to predict the adsorption efficiency all computations were calculated using the EXCEL 97, and the modular artificial neural networks and a Neuro-fuzzy were created with an NN toolbox by using computer codes written in MATLAB mathematical software. The two models and its parameters variation were also determined based on the (ARE), (AARE), (MSE), (RMSE) of the training and prediction set. Before starting the training (learning) process, the values of input neurons were normalized in the 0–1 range to prevent some training pathologies and ensures that the network is not saturated by large values of the weight. To measure the capability of the model for the prediction of unseen experiments which were not used for training, we have 50 experimental runs that randomly split into training, validation and test sets (38-6-6). All samples were normalized.

2.4.1. Artificial neural network (ANN)

A three-layer, an input layer with five neurons (dye concentration, concentration, solution pH, contact time, and adsorbent dose), a hidden layer with seven different modes and an output layer with two neurons (5-7-2), is established [29,30]. The most common network is the back-propagation (BP-ANN) which is a first order gradient descent technique to training algorithm for modeling the experimental data. It is a descent algorithm to minimizing the error at each repetition. This network the weights are adjusted by the algorithm so that the error is reduced along a descent direction. Amongst the different back-propagation (BP) algorithms, we have used The Marquardt–Levenberg learning algorithm. Herein, for all data sets in ANN, the log-sigmoid transfer function (log sig) at hidden layer with five neurons in the first layer and a linear transfer function in the output node were used for the simulation and prediction of dye and Cu (II) removal.

2.4.2. Modeling using adaptive neuro-fuzzy inference systems (ANFIS)

The ANFIS system was applied in order to find a suitable model between the observed inputs and target values, correctly. It is concluding five layers likes, which perform different actions, a fuzzy layer, a product layer, a normalized layer, a defuzzy layer and a total output layer [31–33]. Here, linear and nonlinear parameters were calculated in ANFIS 204, and 340, respectively. The fuzzy inference systems are composed of 34 rules.

2.5. Adsorption experiments and analytical methods

The adsorption process was derived from batch experiments. Following the batch procedure, Batch experiments were conducted in 250 ml glass-stoppered Erlenmeyer flasks reactor containing test solutions at the desired level of dye concentration, Cu concentration, solution pH, contact time, and adsorbent dose at room temperature (22 ± 2 °C). In each test, Quantities of a solution containing a given concentration of Solutions (BR46 dye and Cu, simultaneously) was transferred into the reactor. The initial pH of the solution was adjusted with dilute 0.1 M HCl or NaOH in order to maintain constant pH throughout the experiment. The required weight of adsorbent was added in the specified dosage, and time-course was agitated at 100 rpm using a mini table shaker. Then, Solid/liquid phases were separated by centrifugation at 3000 rpm for 10 min (Shimifan Company, Tehran, Iran).

2.6. Analysis of the samples

The concentration of, Basic Red 46 in the supernatant solution before after and adsorption was determined using a double beam UV/vis spectrophotometer (T80/T80+) at $\lambda_{max} = 530$ nm [8]. Determination of residual concentrations of Cu after adsorption was used by atomic absorption spectroscopy (Varian-AA240). A hydrophilic PTFE syringe filter (0.22 μm pore size) was used for filtering the final suspension. The Removal efficiency of Basic Red 46 and Cu was calculated by the following equation:

The supernatant was analyzed for residual concentrations of dye and metal ion using spectrophotometer UV/VIS (T80/T80+) at $\lambda_{max} = 530$ nm and atomic absorption spectroscopy (Varian-AA240), respectively. Also, a hydrophilic PTFE syringe filter (0.22 μm) was used for the total removal of adsorbent particles from the solution before measuring the copper concentration. After the determination of the concentration of dye and a metal ion, removal efficiency of Basic Red 46 and Cu was calculated by the following equation [34,35]:

$$\%R = \frac{C_0 - C_e}{C_0} \times 100 \quad (1)$$

where C_0 and C_e ($mg.L^{-1}$) are initial and equilibrium concentrations of dye and a metal ion, respectively; V(L) is the sample volume, and w (g) is the mass of adsorbent. Adsorbate uptake on adsorbent at equilibrium state was calculated as Follow [36]:

$$q_e = \frac{(C_0 - C_e) \times V}{w} \quad (2)$$

3. Results and discussion

3.1. Characterization of adsorbent

The FTIR study of the adsorbent of sawdust after simultaneous removal of dye and heavy metal demonstrated significant peak at (3362, 3419 cm^{-1}), and 2913 cm^{-1}which were due to the presence of hydroxyl groups (–OH), and asymmetrical stretching vibration of C-H. Also, the peak recorded at 2364 cm^{-1}, 1425, and 1459 cm^{-1} represents the presence of N═C groups [37], the blending vibration CH_3, and scissor vibration CH_2. A previous study by Dolatabadi et al. was proved by the results of this study [10]. According to Fig. 1, the stretching vibration of C-O is related to peaks between 1000 and 1500 cm^{-1}. The sulfates in the composition of CuSO4 appear at 1140-1200 cm^{-1}. It is remarkable to mention carboxylic groups C═O have an absorbance in the region of 1650–1900 cm^{-1} while the bands of 400–650 cm^{-1} propose the formation of heavy metal coupled with oxygen (M − O). This

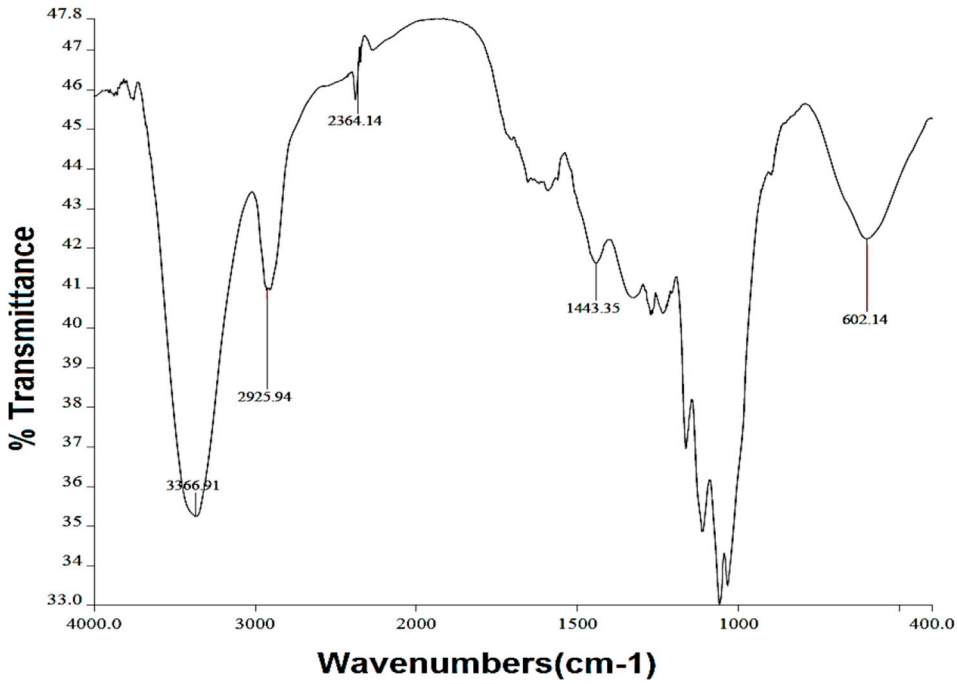

Fig. 1. FT-IR spectra of modified sawdust.

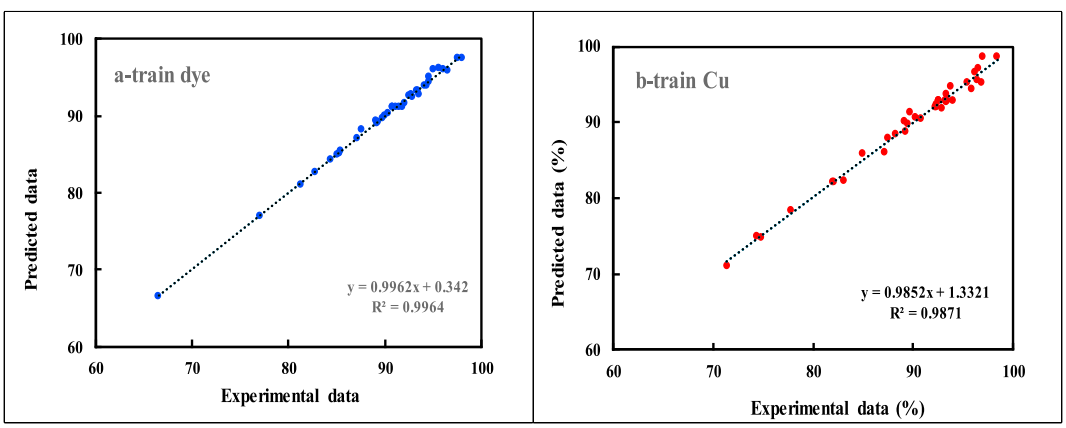

Fig. 2. Comparison of the experimental and predicted results for (a) dye and (b) Cu removal in ANN.

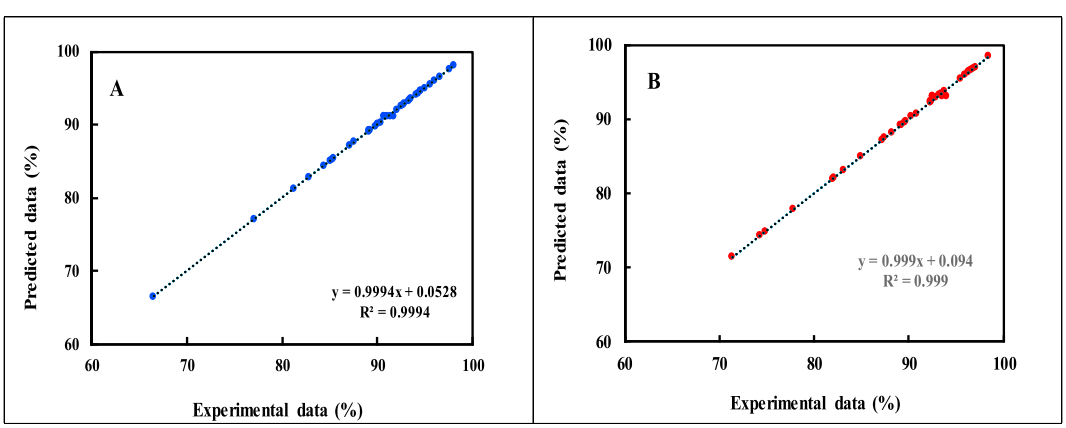

Fig. 3. Comparison of the experimental and predicted results for (a) dye and (b) cu removal in ANFIS.

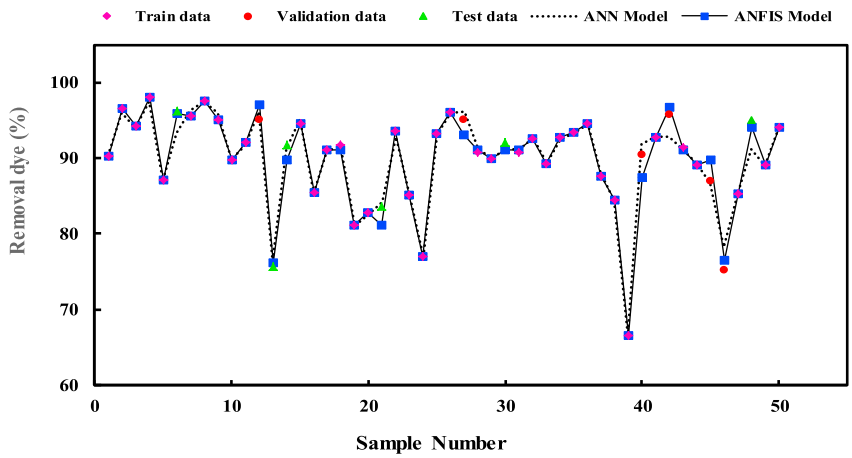

Fig. 4. Comparison experimental data and predicted data by ANN and NFIS for metal ion (Cu) removal.

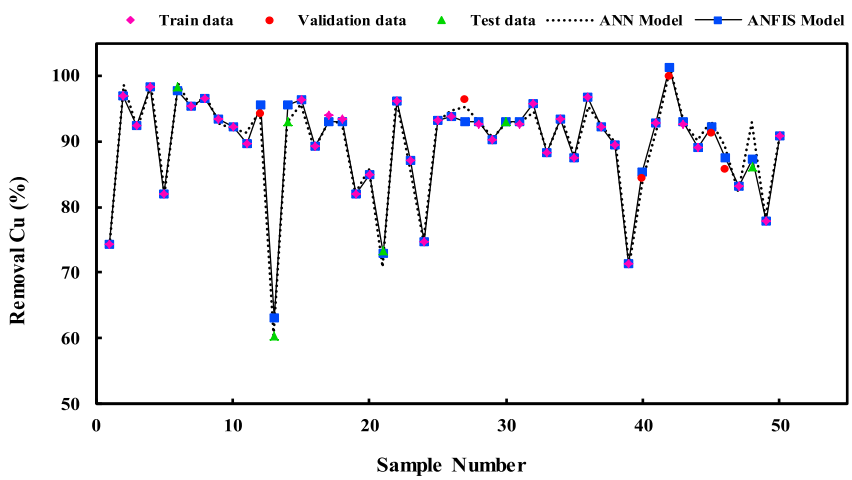

Fig. 5. Comparison experimental data and predicted data by ANN and NFIS for metal ion (Cu) removal.

result represents that the functional groups have been activated by metal adsorbed on the sawdust surface. Due to basic changes in the bands of 3200–3600 cm^{-1}, the OH group was considered as the most effective group in the adsorption process.

In this study Zero electrical charges on the adsorbent surface (pH_{pzc}) was obtained 2.8, implying that the net surface charge is negative at pH medium above 2.8, positive at pH medium below 2.8, and neutral at pH 2.8. In the present study, pH_{pzc} value is equivalent to 2.8. It implies that the surface charge is positive at pH values lower than 2.8, neutral at pH = 2.8 and negative at pH values higher than 2.8.

Table 2
The data used for ANN and ANFIS models by back propagation.

ANN information		ANFIS information	
Characteristic	Value	Characteristic	Value
Number of input nodes	5	Number of nodes	416
Optimum No of neurons in first layer	5	Number of linear parameters	204
Number of output nodes	2	Number of nonlinear parameters	340
Learning rule	Trainlm (LM)	Total number of parameters	50
Number of epochs	25	Number of training data pairs	38
Error goal	0	Number of checking data pairs	6
Mu	10e-5	Number of testing data pairs	6
		Number of fuzzy rules	34

3.2. Explanation of variables

Input variables of models were as follows: initial dye concentration 5–50 mg L^{-1} initial Cu concentration 1–10 mg L^{-1}, pH 2–10, contact time 5–90 min, and adsorbent dosage 1–8 g L^{-1}. The ANN having seven Different modes in the hidden layer was selected for modeling the dye and metal ion adsorption by using sawdust. In Table 1 the best R^2 value for BPNN with a different structure for dye and metal ion adsorption was obtained at 0.96, 0.99 for dye and Cu removal, respectively.

3.3. ANN modeling results

As seen as in Fig. 2 (a,b) neural network prediction of dye and metal ion removal percentage versus their experimental values for the ANN model were studied in training dataset. 38 sets of data selected randomly were used as train in this model. The determination coefficients were obtained at 0.99, 0.98 for dye and metal ion during training, respectively.

Note that the coefficient of determination (R^2) can be calculated as displayed follow [38–40]:

Table 3
Uncertainty measuring parameters for different ANN and ANFIS models for Dye removal.

variable	ARE		AARE		MSE		RMSE	
	ANN	ANFIS	ANN	ANFIS	ANN	ANFIS	ANN	ANFIS
Train	0.0	0.0	0.003	0.003	0.13	0.021	0.247	0.047
Test	0.001	0.012	0.28	0.017	9.661	2.073	2.44	1.183
Total	−0.001	0.0	0.008	0.006	1.696	0.838	0.676	0.426

Table 4
Comparison of ANN and ANFIS model for Cu removal.

variable	ARE		AARE		MSE		RMSE	
	ANN	ANFIS	ANN	ANFIS	ANN	ANFIS	ANN	ANFIS
Train	0.0	0.0	0.007	0.003	0.564	0.046	0.608	0.081
Test	−0.066	−0.008	0.78	0.017	81.815	2.462	5.143	1.016
Total	−0.009	−0.002	0.16	0.006	10.63	0.707	1.248	0.353

$$R^2 = \frac{\left(\sum_{i=1}^{n}(y_{di} - \overline{y}_{di})(y_i - \overline{y}_i)\right)^2}{\sum_{i=1}^{n}(y_{di} - \overline{y}_{di})^2(y_i - \overline{y}_i)^2} \tag{3}$$

Where y_i is the predicted value, y_{di} is the actual value, Q_p is the predicted value, n is the number of observations and the symbol − is the average of the related values.

3.4. ANFIS modeling results

The actual data versus the prediction values of dye and Cu (II) removal by ANFIS model are presented in Fig. 3.a, b The ANFIS model with the coefficient of determination $R^2 = 0.999$ which indicated good agreement in dye and Cu removal by sawdust in aqueous solution.

3.5. Comparison of simulation results from AAN and ANFIS

The parameters and information based on the ANN and ANFIS models during training are reported in Table 2. Among the different training algorithm, Levenberg-Marquardt (LM) was found to have the best performance.

The graphical comparison between the simulation results for dye and metal ion removal using developed AAN and ANFIS models is illustrated in Figs. 4 and 5. According to this result, it was observed that there is a good agreement between the simulation results and the experimental data (actual data). The result revealed that both of the models, satisfactorily, shown good $R2$ and an excellent fitness of predicted and experimental values with laboratory measurements. Also, this showed that the accuracy of the ANN and ANFIS models is acceptable. Thus, the ANN and ANFIS system can be effectively used for predicting model of simultaneous removal of dye and metal ion from aqueous solution. It is clear that the neural network and Neuro-Fuzzy model shows good fitness between predicted and experimental values with excellent R^2 in this work.

3.6. Error analysis

The convergence of solution was obtained for a minimum error when experimental data are fitted using the models. Although it may increase performance it can cause a low stability of the response. An error tolerance must be defined to obtain the model. Thus, to evaluate the prediction performance of model, using the statistical standards, the average relative error (ARE), the absolute average relative error (AARE), Mean square error (MSE), and Root Mean square error (RMSE) were computed for dye and Cu removal in both models (ANN and ANFIS) in Tables 3 and 4, respectively. The values of ARE, AARE, MSE, and RMSE are calculated as follows [41–43]:

$$ARE = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{X_{experimental(i)} - X_{calculated(i)}}{X_{experimental(i)}}\right) \tag{4}$$

$$AARE = \frac{1}{N}\sum_{i=1}^{N}\left(\left|\frac{X_{experimental(i)} - X_{calculated(i)}}{X_{experimental(i)}}\right|\right) \tag{5}$$

$$MSE = \frac{\sum_{i=1}^{n}\left(X_{experimental(i)} - X_{calculated(i)}\right)^2}{N} \tag{6}$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}\left(X_{experimental(i)} - X_{calculated(i)}\right)^2}{N}} \tag{7}$$

All the train and test data investigated with ANN and ANFIS models. It is obvious that the simulation results are very satisfactory agreement with ANFIS than ANN model.

4. Conclusion

The main objective of the present study was to investigate the abilities of an artificial neural network (ANN), and adaptive-network-based fuzzy inference system (ANFIS) models utilization in predicting the percentage of dye and Cu (II) removal by sawdust from the aqueous solution. According to the reported results, it is evidenced that ANN and ANFIS is a promising predicting technique that can be used effectively with satisfactory accuracy for the prediction of the simultaneous dye and Cu (II) removal from the aqueous solution. In almost all the train and test data, the performance of the both of the models in Prediction and simulation was measured using the (ARE), (AARE), (MSE), (RMSE) and the correlation Coefficients R^2 values. The lower value of the statistical parameter shows better performance both of the model. Here, a good regression analysis with the R^2 in the range of 0.98–0.99 for dye and Cu both of the models were obtained.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.chemolab.2018.07.012.

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