

Prediction of the continuous cadmium removal efficiency from aqueous solution by the packed-bed column using GMDH and ANFIS models

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ABSTRACT

In the present study, the performance of a packed-bed column filled with *Sargassum angustifolium* brown seaweed in cadmium ions removal from the aqueous solution was predicted using the group method of data handling (GMDH), artificial neural network (ANN), and adaptive neuro-fuzzy inference system (ANFIS) models. During the modeling, various values of column bed height and feed flow rate to the column were considered as the model inputs while the adsorption efficiency was predicted as the output parameter of the model. Comparison between the predictions and the real experimental data indicated that both of the applied models had a great and almost similar performance ($R^2 > 0.99$) in the prediction of cadmium uptake from the aqueous solution. Moreover, mean squared error values were 0.0006 and 0.0003 for GMDH and ANFIS, respectively. According to the results, the two proposed models showed a great potential in the prediction of cadmium adsorption from aqueous solutions using a packed-bed column filled with *S. angustifolium*.

Keywords: Artificial neural network-group method of data handling; Adaptive neuro-fuzzy inference system; Packed-bed column; *Sargassum angustifolium*; Adsorption; Cadmium

1. Introduction

Water is one of the most significant resources which has been severely exposed to various pollutants. Water contamination occurs when the value of one or more substances in water exceeds the standard limit, which can be harmful to humans and animals [1]. Heavy metal ions can cause water pollution through human activities like mining, tanning, metal industries, steel production, discharge of municipal sewage, and usage of agricultural chemical fertilizers [1–3]. Due to the acute and drastic effects of these pollutants on human health, heavy metals such as cadmium are considered as non-biodegradable highly toxic elements even at low traces. Exposure to low concentrations of cadmium (30–50 µg/d) can lead to bone diseases,

cancer, kidney failure, and hypertension [4–6]. The maximum allowable cadmium concentrations in wastewater and drinking water are 0.1 and 0.005 mg/L, respectively. Considering the problems associated with the inappropriate discharge of such chemical pollutants into the environment, the treatment of effluents containing heavy metal ions before the discharge into the environment or wastewater network seems essential [7].

Environmental researchers are always trying to find an easy, effective, economical, and environmentally friendly solution to remove heavy metals from effluents [8–11]. Among these methods, due to their simple design, ease of operation, applicability to a variety of toxic metals, and adsorbent regeneration capability, adsorption is the safest choice [12]. It describes the removal of heavy metals from the

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aqueous solution by a mechanism attributed to the ligands found in the cellular wall of biomolecules. Additionally, it is highly efficient, especially at low metal concentrations (below 100 mg L⁻¹) [13,14]. Several applications of packed-bed columns to remove heavy metal ions from aqueous solutions have been observed due to their continuous operation and high removal rates [15]. The design of such columns is simple and also they are regenerative, free of sludge, easy to operate, and have high efficiency and adsorption capacity [16–18]. High concentration flow of metallic ions in the influent of the column is exposed to the new biosorbent, where the highest driving force, which is formed between the concentration of ions in water and those on the biomass surface (0 mg/L), leads to complete removal of metallic ions [19,20]. Various biomasses such as algae and brown and green seaweeds have been used in packed-bed columns for removing heavy metals [21–34]. Among them, seaweeds have received great attention due to their low price, low sensitivity to environmental factors and impurities, adsorbent regeneration capability, contaminant recovery, and high adsorption efficiency. It has also been confirmed that the heavy metal ion adsorption capacity of brown seaweed, such as *Sargassum* is much higher than others [35].

Adsorption of heavy metal ions from wastewaters is a very complex process because of the influence of several operating parameters which leads to nonlinear relationships between process inputs and outputs; as a result, it is difficult to model such systems using conventional mathematical models [36,37]. Artificial neural network (ANN) and adaptive neuro-fuzzy inference system (ANFIS) are appropriate alternatives due to the high potentials in modeling nonlinear systems. Over the past decades, ANN and ANFIS models have been employed in various fields of engineering. Singh et al. [36] predicted cadmium adsorption by hematite adsorbent using ANN and ANFIS models. In 2014, Ahmad et al. [38] studied the usage of ANN model to predict the adsorption capacity of immobilized *Bacillus* adsorbent for removal of cadmium ions from the aqueous solution using the Levenberg Marquardt training algorithm. In another work, Nasr et al. [39] investigated the prediction of rice straw adsorption efficiency for removal of cadmium ions using the ANFIS model.

In this study, the ANFIS and a new ANN model called the group method of data handling (GMDH) were applied to model the performance of a packed-bed column in the removal of Cd ions from aqueous solutions using the adsorbent produced from *Sargassum angustifolium* brown seaweed for the first time. In order to do this, the experimental data of previously published research by Jafari and Jamali [40] were used. The predicted results of both models were compared with the experimental data to assess the performance of the two proposed models.

2. Materials and methods

2.1. Experiments

As mentioned, the present study aimed to predict the performance of a packed-bed column filled with *S. angustifolium* brown seaweed in cadmium removal from aqueous solutions using GMDH ANN and ANFIS models

based on the previously reported experimental data [40]. The seaweed collection procedure, adsorbent preparation, column features, and the adsorption procedure have been reported completely in Jafari and Jamali's work [40]. The internal diameter and height of the column were 2.5 and 20 cm, respectively. In order to provide a uniform solution flow into the column and pack the biomass, a 3 cm height of glass beads was placed at the top and bottom of the bed. Additionally, a stainless steel sieve was placed between the glass beads and bed to prevent biomass loss. A peristaltic pump was used to generate the constant flow rate of the solution from the bottom of the column. After preparing the feed cadmium solution by dissolving the required quantity of cadmium salt in distilled water, pH of the solution was adjusted to 6 using NaOH 0.1 M and HNO₃ 0.1 M. According to the experiments, the effects of bed height (2.6, 4.9, and 7.5 cm) and feed flow rates values (15, 20, and 30 mL min⁻¹) on the column adsorption performance were investigated. The sampling was done at the column output at different time intervals and then the samples were analyzed using a flameless atomic absorption spectrophotometer (PG Instruments, AA500, England) to measure the cadmium residual concentration [40].

2.2. Methods

2.2.1. GMDH neural network

GMDH neural networks are a type of GMDH algorithms which are expressed in the form of network structures. This type of neural network is a self-organized and unilateral network that consists of several layers, each one is composed of several neurons. All of the neurons have a similar structure. Certain and constant weight (w) values are specified for neurons based on solving orthogonal equations. The prominent feature of these networks is that the neurons of the previous layer are the generators of new neurons. In order to avoid the divergence of the network, a number of the newly generated neurons must be eliminated which are called dead neurons. One of the important issues in the design of multilayer ANNs is their internal structure, including the number of weights and their initial values as well as the proper selection of the stimulation function of each neuron to generate a suitable and ideal mapping between the input and output data. The purpose of GMDH neural networks is to prevent the increase of network divergence and associate the shape and structure of the network to one or more numerical parameters in a way that the network structure is varied by changing such parameters [41,42].

GMDH uses a second-order polynomial transfer function to determine an analytical function whose coefficients are achieved from regression coefficients. The estimation of \hat{f} function should be performed in a way that it would be used instead of the real f function. For an input vector $[X = (x_1, x_2, x_3, \dots, x_n)]$, the value of estimated parameter (\hat{y}) should be close to the real value of y . In order to observe M pairs of data, the objective vector is shown by Eq. (1):

$$y_i = f(x_{i1}, x_{i2}, x_{i3}, \dots, x_{in}) \quad (i = 1, 2, 3, \dots, M) \quad (1)$$

During the training of the GMDH network, the estimates of the objective parameter (\hat{f}_i) using the input vector are presented as follows:

$$\hat{y}_i = \hat{f}(x_{i1}, x_{i2}, x_{i3}, \dots, x_{in}) \quad (i = 1, 2, 3, \dots, M) \quad (2)$$

The proposed model in this research should minimize the deviation between the observed values and the real data. As a consequence, the defined objective function of the problem, which is the square of differences between estimated and observed outputs and it is shown in Eq. (3) should be minimized by GMDH:

$$D = \sum_{i=1}^M (\hat{f}(x_{i1}, x_{i2}, x_{i3}, \dots, x_{in}) - \hat{y}_i)^2 \rightarrow \text{Min} \quad (3)$$

Volterra series functions are usually applied in modeling complex systems with several inputs and one output:

$$\hat{y} = a_0 + \sum_{i=1}^n a_i x_i + \sum_{i=1}^n \sum_{j=1}^n a_{ij} x_i x_j + \sum_{i=1}^n \sum_{j=1}^n \sum_{k=1}^n a_{ijk} x_i x_j x_k + \dots \quad (4)$$

In basic methods of GMDH algorithm, all of binary compounds (neurons) are generated by n input variables and unknown coefficients of neurons are achieved by minimum squares method. Therefore, $\binom{n}{2} = \frac{n(n-1)}{2}$ neurons are generated in the second layer which is shown as Eq. (5):

$$\left\{ (y_i, x_{ip}, x_{iq}) \mid (i = 1, 2, \dots, M) \quad \& \quad p, q \in (1, 2, \dots, M) \right\} \quad (5)$$

For each ternary M rows, the second-order form of Eq. (5) is used. These equations can be expressed in the matrix form of Eq. (6):

$$A_a = Y \quad (6)$$

where A is the unknown coefficients vector of the second-order equation [Eq. (5)]:

$$a = \{a_0, a_1, \dots, a_5\} \quad (7)$$

And

$$Y = \{y_1, y_2, y_3, \dots, y_M\}^T \quad (8)$$

Considering the input vector values and the function form, it can be observed that:

$$A = \begin{bmatrix} 1 & x_{1p} & x_{1q} & x_{1p}^2 & x_{1q}^2 & x_{1p} & x_{1q} \\ 1 & x_{2p} & x_{2q} & x_{2p}^2 & x_{2q}^2 & x_{2p} & x_{2q} \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ 1 & x_{Mp} & x_{Mq} & x_{Mp}^2 & x_{Mq}^2 & x_{Mp} & x_{Mq} \end{bmatrix} \quad (9)$$

Using the minimum squares method of multiple-regression analysis, the solution to the equation is given by:

$$a = (A^T A)^{-1} A^T Y \quad (10)$$

The current equation gives the coefficients of correlation 5 for all of M ternary sets [43–46].

2.2.2. Adaptive neuro-fuzzy inference system

A fuzzy system is based on fuzzy if/then rules which cannot be analyzed by classical probability theories. Fuzzy logic aims to extract accurate results using a set of rules defined by experts. Moreover, neural networks are capable of being trained and can determine network parameters using the observed data to achieve the desired output for an arbitrary input value. However, contrary to fuzzy systems, neural networks are not capable of using human knowledge and interference based on linguistic expressions [47,48].

A sugeno-type fuzzy system is the base of the ANFIS model. This model consists of five layers:

Input layer (input nodes): each node of this layer generates the membership values which belongs to each of the fuzzy sets using the membership function:

$$\begin{cases} O_{1,i} = \mu_{A_i}(x) & \text{for } i = 1, 2 \\ O_{1,i} = \mu_{B_{i-2}}(y) & \text{for } i = 3, 4 \end{cases} \quad (11)$$

where x and y are non-fuzzy inputs to node i , and A_i and B_i are linguistic labels, which are specified by membership functions μ_{A_i} and μ_{B_i} ; these functions are denoted by symbol O . The parameters of these membership functions, which are known as the primitive parameters of the layers, must be specified. For example, one of the membership functions used in the models is the bell-shaped function, which is as follows:

$$\mu_A(x) = \frac{1}{1 + \left| \frac{x - c_i}{a_i} \right|^{2b_i}} \quad (12)$$

where a_p , b_p , and c_i are the basic parameters of the model which should be trained.

Second layer: in this layer each neuron is constant and the “And” operator should be applied to obtain the output that represents the antecedent part of that rule which forms the output function of the rule. Therefore, the O_{2k} outputs of this layer are the result of the product of the degrees associated with the first layer:

$$O_{2,i} = w_i = \mu_{A_i}(x) \cdot \mu_{B_i}(y) \quad i = 1, 2 \quad (13)$$

Third layer: the main purpose of the third layer is to determine the ratio of each output of the i th rule to the sum of all the outputs of the rules; as a result, w is obtained as a standard output:

$$O_{3,i} = \bar{w}_i = \frac{w_i}{w_1 + w_2} \quad (14)$$

Fourth layer (result nodes): in this layer, the output of each node is presented by Eq. (15):

$$O_{3,i} = \bar{w}_i f_i = \bar{w}_i (p_i x + q_i y + r_i) \quad (15)$$

where w_i is the output of the i th node of the previous layer and p_i , q_i , and r_i are the coefficients of this linear combination. *Fifth layer (output nodes):* this layer calculates a single output node by summation of all the input signals. Therefore, in this layer the results of each fuzzy rule are transformed into non-fuzzy outputs by the defuzzification process:

$$O_{5,i} = \sum_i \bar{w}_i f_i = \frac{\sum_i w_i f_i}{\sum_i w_i} \quad (16)$$

This network is trained based on supervised learning. Therefore, the objective is to train adaptive neuro fuzzy networks which can estimate the undefined functions achieved from the training data and find the exact values for the above parameters. The distinguishing feature of ANFIS is to provide the training algorithm, gradient descent, and least squares to modify the parameters [49,50].

Subtractive clustering (SC) is a method that divides an input space into n subdivided specific areas through analyzing n -dimensional input data to create clusters. This method divides the input space into appropriate clusters even if the user does not specify the number of clusters. The radius of the cluster is the range of influence from the center of the cluster when the input space is considered as a unit hypercube, by the user. The radius of the cluster has a value between 0 and 1. One of the significant issues in using SC is to determine the appropriate radius for the estimation of the number of clusters. Therefore, specifying a low value for the radius results in considerably high number of clusters; as a result, there will be too many rules. On the other hand, high radius values leads to unacceptable results. It should be mentioned that this method uses constant output [51,52].

This structure extracts a series of fuzzy rules to model the training matrix behavior. In order to extract the rules, subtracting clustering algorithm should be initially used to determine the rules, initial values of membership functions, and the number of membership functions, then linear least-square estimates were applied to find the equation of each rule. Decreasing clustering algorithm divides the dimension of input and output data between a number of clusters based on the impact range of the center of clusters. A membership function is specified for each cluster in each dimension of input and output data. The input and output membership functions are Gaussian and linear, respectively. Additionally, the number of output membership functions is 1.

ANFIS is a combination of ANN and fuzzy inference systems (FIS). This combination has advantages that can overshadow the deficiencies of the two methods. ANFIS model can be trained without the need for adequate and special knowledge for a fuzzy logic model. The outstanding feature of ANFIS is having both numerical and linguistic knowledge. This method uses ANN's capability in the

classification of data and identifying the patterns. Compared to ANN, ANFIS is more clear which leads to lower errors in train, other advantages of ANFIS are the ability to capture the nonlinear structure of a process, adaptation capability, and rapid learning capacity.

GMDH models can also achieve explicit expressions, while ANFIS provides implicit models like black boxes. Such a feature helps the researchers to find the maximum number of important variables. GMDH offers network-compatible displays, while the topology of ANFIS is designed based on trial and error procedure.

3. Results and discussion

3.1. Modeling by GMDH and ANFIS

In order to model a process by GMDH and ANFIS techniques, a series of input and output data is required. In the current research, variables such as column bed height, time, and feed flow rate are considered as the input parameters while cadmium removal percentage is the network output.

The datasets which are used to design a GMDH or neural fuzzy network system are divided into two categories: training and validation. In the network design, the whole information of the training category is given to the network. In other words, the network receives both inputs and outputs of the training set and adjusts its parameters based on this information during the training step. By contrast, in the testing step, only the inputs are given to the network and the network has no previous knowledge about such data. Furthermore, validation step aims to prevent the network from memorizing the data, instead of learning. In other words, this step is a limiting one for neural and neural-fuzzy networks, since it stops the network from memorizing the data. In this work, 75% of the data were selected randomly for training and remained 25% were used for validation. Furthermore, mean squared error (MSE) and mean absolute error (MAE) were calculated to validate the performance of the proposed models.

It is clear that the lower the error of the training data set, the higher the performance of the training step. However, this is not always true; in many cases, it has been observed that the training error is moderately low, while the network output is not suitable, since the network is memorizing rather than learning. This problem can be solved by using the validation dataset.

In this research, MATLAB software was used for the modeling of the performance of a packed-bed column filled with brown seaweed *S. angustifolium* in cadmium ions uptake from the aqueous solution. Moreover, the designed GMDH neural network consisted of three layers: an input layer, a hidden layer, and an output layer. The only difference between these layers was in the number of neurons in each one. In order to define the number of neurons in the hidden layer, the error was calculated for different networks and finally, the best network with the lowest training error was selected. The algorithm used for GMDH neural network is shown in Fig. 1.

During the modeling using ANFIS, the general structure of the model including input, output, and model functions should be determined initially. Three methods including

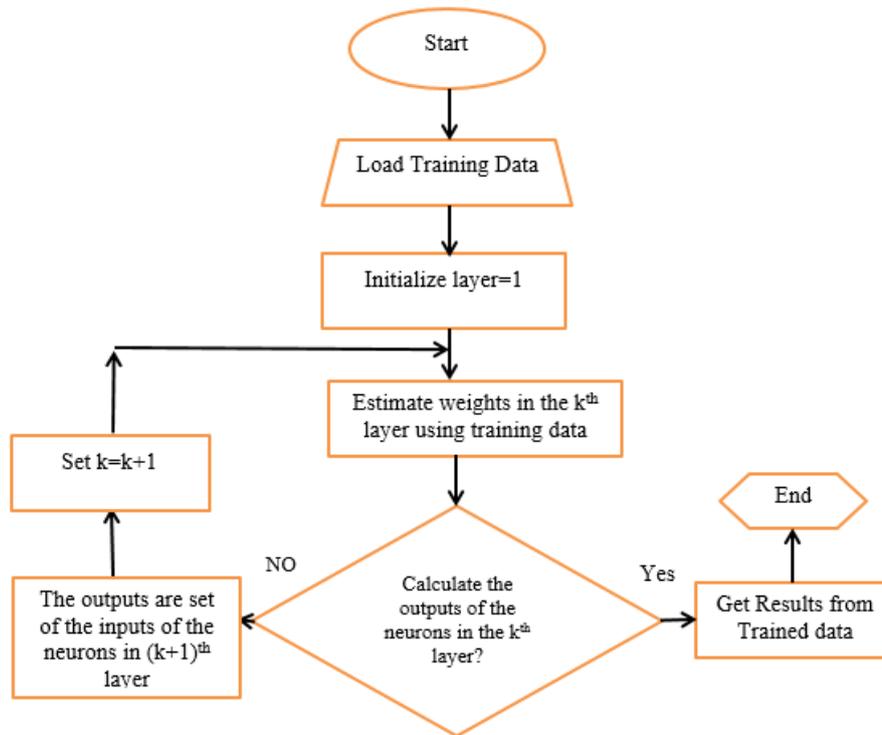


Fig. 1. Block diagram representation of training algorithm for GMDH [53].

grid partitioning, fuzzy C-mean clustering, and subtractive clustering are usually applied in modeling by neuro-fuzzy systems. The major difference between these two methods is in the determination of the fuzzy membership functions. In the network discretization, the type and number of input data vector membership functions are selected by the user, while in the subtractive clustering technique the type of membership functions is determined by the neuro-fuzzy inference model based on the input data vector properties and their classifications. It should be noted that in this work, the subtractive clustering method was utilized to predict the efficiency of cadmium removal from the aqueous solution by the column filled with brown seaweed *S. angustifolium*. A hybrid method was used to train the model, which was a combination of neural network and fuzzy methods. The used ANFIS algorithm is shown in Fig. 2.

3.2. Modeling results

In the present research, GMDH and ANFIS models were used to predict the performance of a packed-bed column filled with *S. angustifolium* brown seaweed in the removal of cadmium ions from the aqueous solution. The bed height, time, and flow rate into the column were considered as the input parameters and cadmium adsorption efficiency was the output variable of the models. The data were normalized between 0 and 1 before modeling to increase the accuracy of the numerical calculations and also to achieve a better output. The statistical information of the data used in this study is reported in Table 1.

As shown in Fig. 3, the majority of the data were in the vicinity of the $Y = X$ line, therefore, it can be concluded

that the capability of the GMDH neural network model in predicting the outputs was acceptable. Additionally, the calculated correlation coefficients for the training and testing datasets for the current model were 0.9969 and 0.9962, respectively.

The proposed structure of the GMDH neural network generalized for modeling and prediction of C_e/C_i is provided in Fig. 4. In the GMDH model, the number of layers and neurons in the hidden layers, the structure of the model, and other parameters are determined automatically.

Moreover, the information of the proposed ANFIS model in this study is presented in Table 2.

Fig. 5 depicts the data predicted by the ANFIS model vs. the experimental cadmium adsorption data. Considering the reported data, it can be concluded that the proposed ANFIS model was a reliable tool for predicting the performance of the packed-bed column in the adsorption of cadmium ions from the aqueous solution. As it can be observed the R^2 values were higher than 0.99 which denotes the capability of the designed model in the prediction of outputs.

3.3. Comparing the modeling performance of GMDH and ANFIS

Fig. 6 illustrates a visual comparison between the predicted values and the experimental data of cadmium removal for the column bed height of 2.6 cm. Based on this figure, there is an acceptable agreement between the experimental data and values predicted by both of the proposed models. Furthermore, the correlation coefficients of training and test datasets of the ANFIS model were both higher than 0.99.

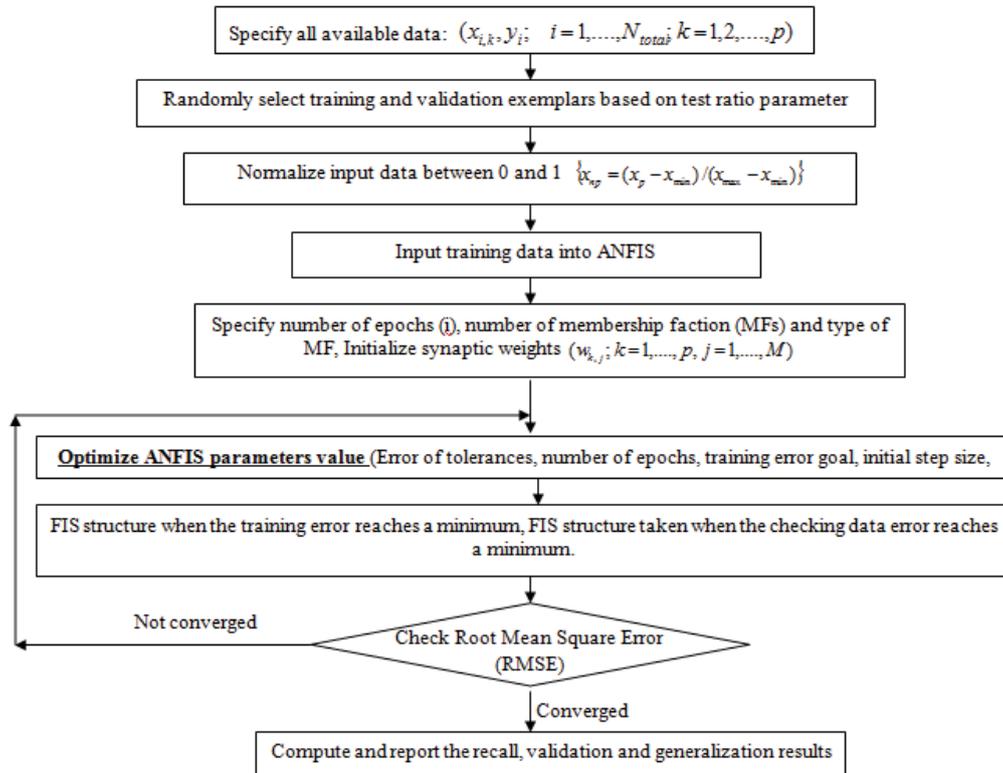


Fig. 2. ANFIS Algorithm [54].

Table 1
Statistical details of the data in the present work

Parameter	Minimum	Maximum	Median	Standard deviation
Column height (cm)	2.6	7.5	2.6	2.0096
Flowrate (mL/min)	15	30	15	5.608
Time (min)	0	2,400	595	554.8125
C _d /C _i (real)	0	0.997615385	0.5077	0.4201
C _d /C _i predicted by GMDH	-0.0205	1.0138	0.5077	0.4212
C _d /C _i predicted by ANFIS	-0.0168	0.9991	0.5268	0.4204

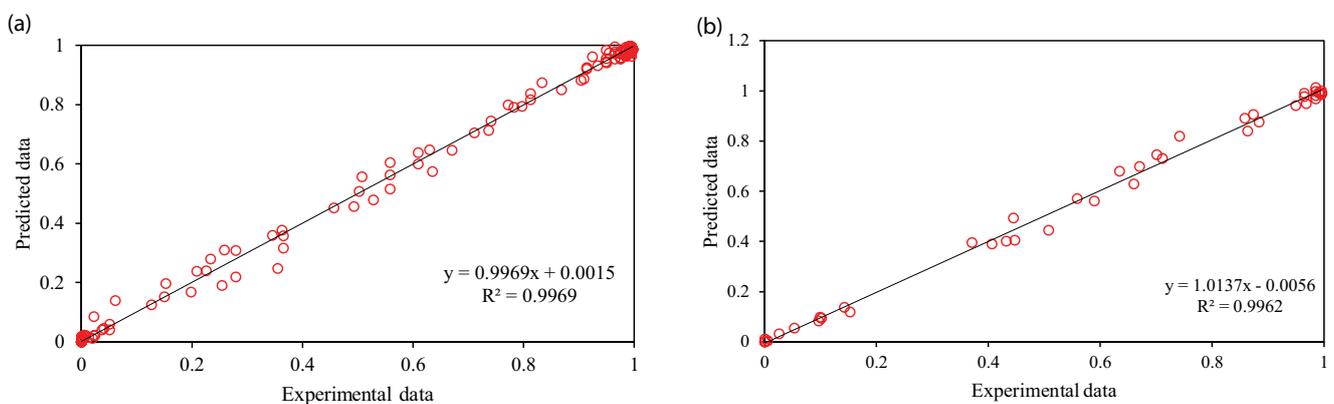


Fig. 3. Comparison between the experimental data and predicted values by GMDH neural network; (a) training and (b) validation datasets.

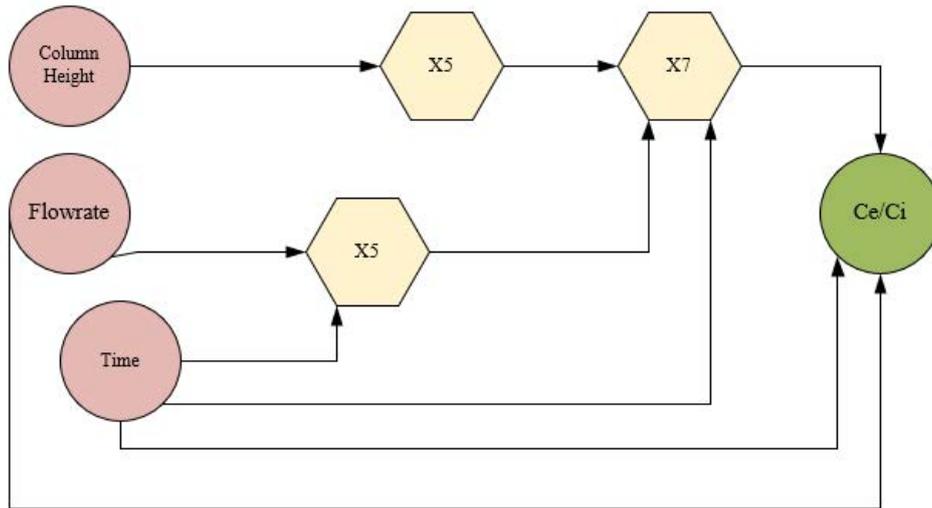


Fig. 4. The structure of the GMDH based mode in C_j/C_i prediction.

Table 2
Technical information of ANFIS model

Characteristics	Value
Number of nodes	158
Number of linear parameters	76
Number of nonlinear parameters	114
Total number of parameters	189
Number of training data pairs	140
Number of testing data pairs	49
Number of fuzzy rules	19
Influence radius	0.3
Number of epochs	1,000
Initial of FIS	Genfis2 (Subtractive clustering)
Accept ratio	0.5
Reject ratio	0.1
Optimization method	Hybrid

Considering this figure, it can be said that an increase in bed height postpones the breakthrough and exhaustion times, increases the critical bed length, and decreases the MTZ slope because of higher surface area and more binding sites for metal sorption. In addition, experimental breakthrough curves are unsymmetrical, which means that experimental data initially split from the horizontal axis fast (a sharp leading edge), although they gradually adjoin the $C_j/C_i = 1$ line at the end of the process. Such a gradual approach is called “tailing”. An increase of the bed height results decrease in the resolution of edges, especially protracted edges. Such behavior can be explained by mass transfer limit or abnormal flow patterns.

Statistical parameters such as MSE, RMSE, MAE, and R^2 were calculated for the proposed models and are presented in Table 3. These data demonstrate that the efficiencies of ANN-GMDH and ANFIS models in modeling the adsorption process were almost identical and their corresponding correlation coefficient values were above 0.99. Additionally, MSE values for the two models were

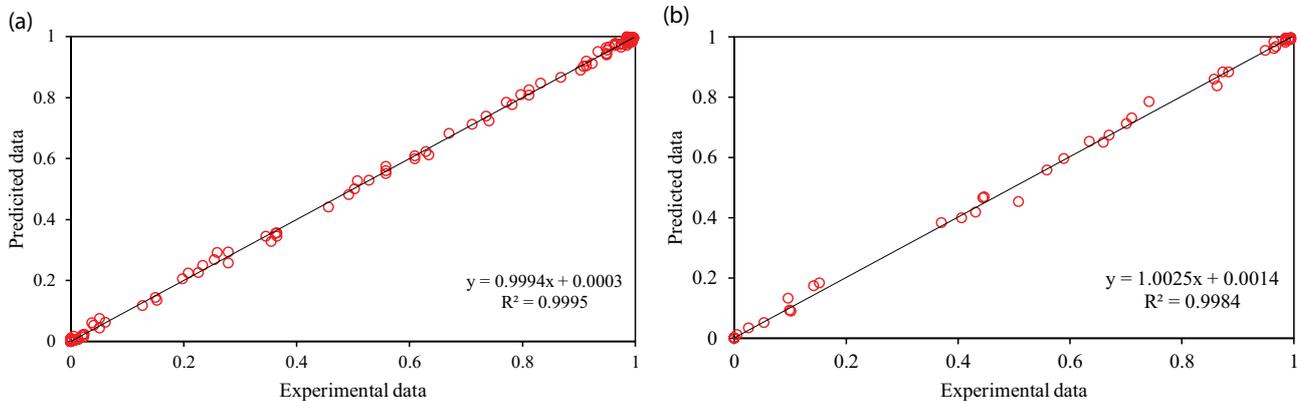


Fig. 5. Comparison between the experimental data and predicted values by the ANFIS; (a) training and (b) validation.

Table 3
Statistical data of ANN-GMDH and ANFIS models

	Method	MAE	MSE	RMSE	R ²
Train	GMDH	0	0.0006	0.0245	0.9969
	ANFIS	0	0.0001	0.0100	0.9995
Test	GMDH	-0.0013	0.0006	0.0245	0.9962
	ANFIS	-0.0027	0.0003	0.0173	0.9984

0.0006 and 0.0003, respectively. Therefore, ANN-GMDH and ANFIS models can be used in modeling the performance of a packed-bed column filled with *S. angustifolium* brown seaweed in cadmium ions uptake from the aqueous solution. It should be noted that the ANFIS model is preferred because of its simple structure and ease of training step compared to the ANN-GMDH model.

Figs. 7 and 8 show the histograms of errors and their distribution for the training and testing data sets. As can

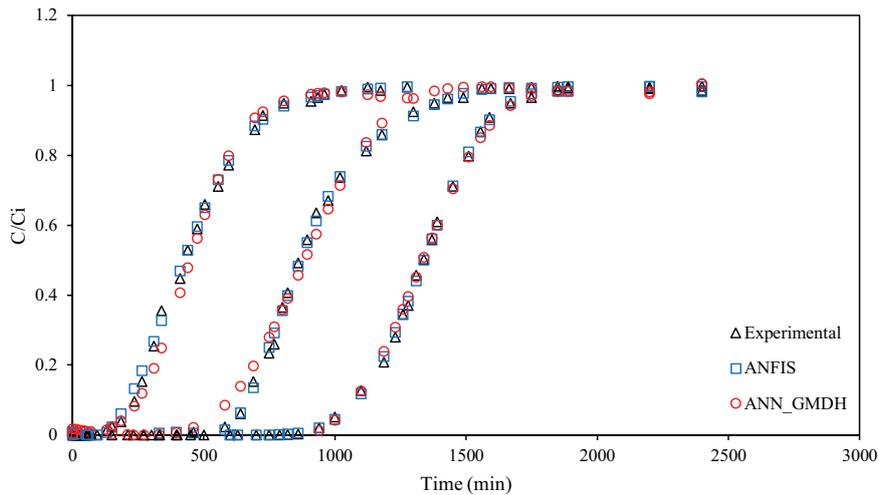


Fig. 6. Comparison between the experimental data and the values predicted by ANN-GMDH and ANFIS for the column height of 2.6 cm.

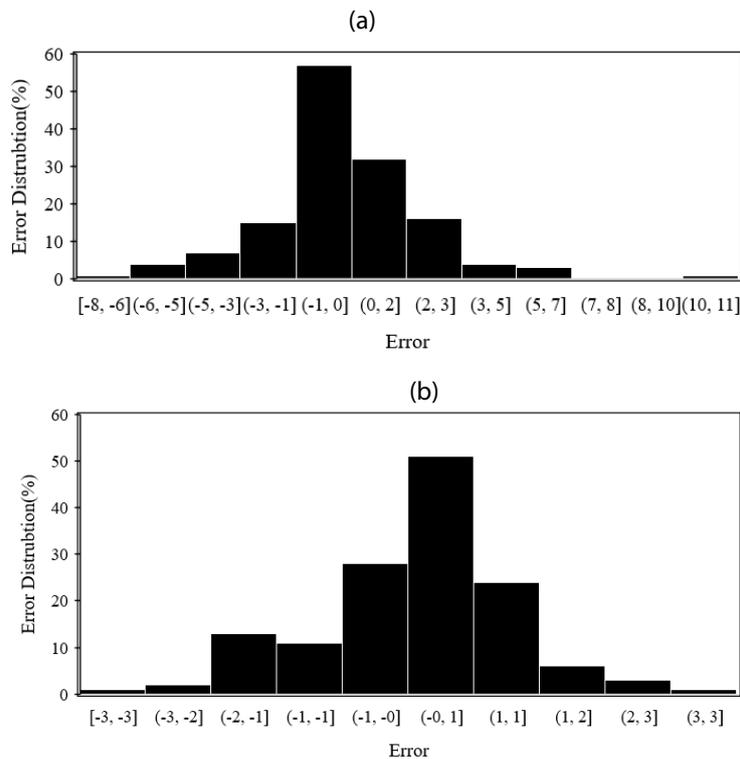


Fig. 7. Histograms for distribution of error values in the training process of (a) ANN-GMDH and (b) ANFIS models.

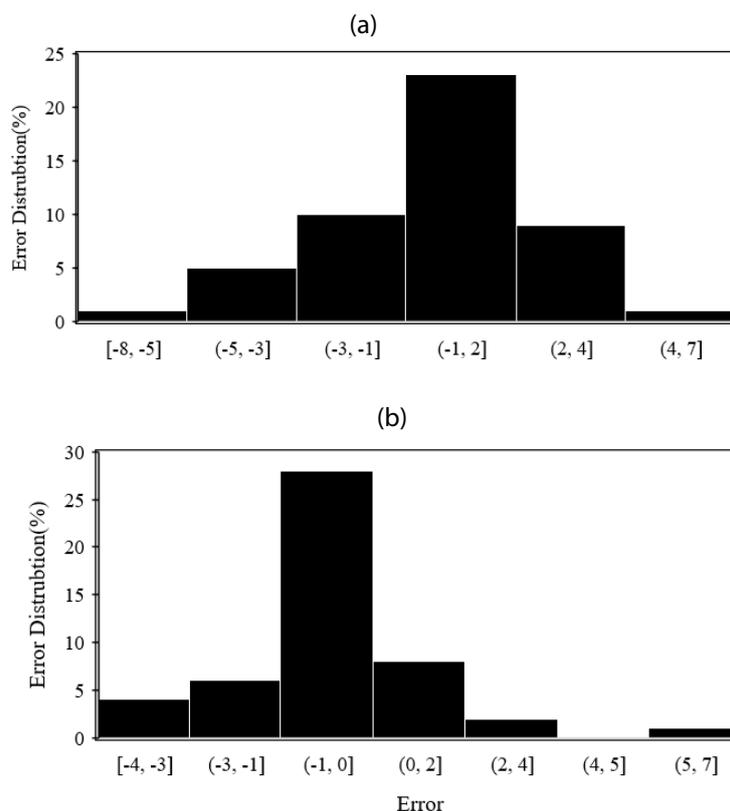


Fig. 8. Histograms for distribution of error values in the testing process of (a) ANN-GMDH and (b) ANFIS models.

be observed, the distribution was higher in near 0% errors. In addition, it decreased for the higher error values.

For the training dataset of the ANFIS model, the error was between -3% and 3% , while in the GMDH model, the error varied between -8% and 11% . Additionally, the highest frequency in both models was -1 and 1 , indicating the agreement between the experimental and predicted data. However, in the testing dataset of the ANFIS model, the error was in the range of -4% to 7% , while for the GMDH model it was between -8% and 11% . It should be noted that training error values were in these ranges. Comparison between these histograms shows that the distribution of errors of the ANFIS model was in a more normal form than that of ANN-GMDH; as a result, better predictions can be expected from the former.

4. Conclusion

In this paper, ANN-GMDH and ANFIS models were applied to predict cadmium adsorption using a packed-bed column filled with *S. angustifolium* brown seaweed. The input variables were the column bed height, time, and feed flow rate while the output parameter of the models was the cadmium removal efficiency. In the ANFIS model, the subtractive clustering and hybrid training algorithms were used. Comparison between the results of the proposed two methods approved the accuracy of ANN-GMDH and ANFIS for predicting packed-bed column removal efficiency since their correlation coefficient

values were extremely high ($R^2 > 0.99$). Additionally, statistical parameters including MSE, RMSE, and MAE were calculated for all training and testing datasets to evaluate the performance of both models in the prediction and simulation of the output values. The low values of the calculated errors indicated the great performance of the two proposed models. Therefore, it is suggested to use ANN-GMDH and ANFIS models for the prediction of cadmium adsorption from the aqueous solution using a packed-bed column filled with *S. angustifolium* brown seaweed.

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